Concentrations of Proanthocyanidins in Common Foods and Estimations of Normal Consumption[1,2]

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ABSTRACT Proanthocyanidins (PAs) have been shown to have potential health benefits. However, no data exist concerning their dietary intake. Therefore, PAs in common and infant foods from the U.S. were analyzed. On the bases of our data and those from the USDA's Continuing Survey of Food Intakes by Individuals (CSFII) of 1994-1996, the mean daily intake of PAs in the U.S. population (>2 y old) was estimated to be 57.7 mg/person. Monomers, dimers, trimers, and those above trimers contribute 7.1, 11.2, 7.8, and 73.9% of total PAs, respectively. The major sources of PAs in the American diet are apples (32.0%), followed by chocolate (17.9%) and grapes (17.8%). The 2- to 5-y-old age group (68.2 mg/person) and men >60 y old (70.8 mg/person) consume more PAs daily than other groups because they consume more fruit. The daily intake of PAs for 4- to 6-mo-old and 6- to 10-mo-old infants was estimated to be 1.3 mg and 26.9 mg, respectively, based on the recommendations of the American Academy of Pediatrics. This study supports the concept that PAs account for a major fraction of the total flavonoids ingested in Western diets. J. Nutr. 134: 613–617, 2004.

KEY WORDS: • *catechin* • *proanthocyanidins* • *tannins* • *foods* • *infant foods*

Proanthocyanidins (PAs),[4] better known as condensed tannins, are oligomeric and polymeric flavan-3-ols. They are ubiquitous and present as the second most abundant natural phenolic after lignin. The flavan-3-ol units are linked mainly through the C4→C8 bond, but the C4→C6 bond also exists (both called B-type). The flavan-3-ol units can also be doubly linked by an additional ether bond between C2→O7 (A-type). The size of PA molecules can be described by their degree of polymerization (DP) (1). Three common flavan-3-ols, which differ in their hydroxylation patterns, are found in PAs. The PAs consisting exclusively of (epi)catechin are procyanidins (PCs). PAs containing (epi)afzelechin or (epi) gallocatechin as subunits are named propelargonidins (PPs) or prodelphinidins (PDs), respectively. PPs and PDs are less common in nature than PCs. They are heterogeneous in their constituent units and coexist with the PCs (1).

PAs are of great interest in nutrition and medicine because of their potent antioxidant capacity and possible protective effects on human health (2). PAs have been suggested to account for a significant fraction of polyphenols ingested in a Western diet because of their ubiquitous existence (2). There have been many studies concerning the content of flavonoids in foods and their daily intake (3–7). However, no data exist concerning the daily intake of oligomeric and polymeric flavan-3-ols because data are lacking on the PA content of foods, in particular. Only recently has an appropriate analytical method been developed for the measurement of PAs (8,9). The objective of the present study was to quantitatively analyze PAs in both common and infant foods and evaluate their daily intake in the U.S. population on the basis of food consumption survey data. This study will establish a PA food database under the USDA National Food and Nutrition Analysis Program.

MATERIALS AND METHODS

Standards. A composite PC oligomer standard containing monomers through decamers was purified from cocoa. A polymer PC fraction with a mean DP of 36.1 was purified from blueberries and used as a polymer standard (9).

Food samples. Foods were sampled from four areas of the United States in two seasons. The samples were colleted and processed in the Department of Biochemistry of Virginia Technical University (Blacksburg, VA). Sorghum samples were provided by Dr. Lloyd Rooney (Texas A&M, College Station, TX). Samples were shipped frozen on dry ice and kept at −70°C before use. The fruits and vegetables were received in the form of freeze-dried powders. The final data were converted to a fresh weight basis using the moisture content of the foods. Nuts, cereals/beans, snacks, and spices were ground without freeze-drying. Beverages were analyzed in their orig-

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[4] Abbreviations used: DP, degree of polymerization; PAs, proanthocyanidins; PCs, procyanidins; PDs, prodelphinidins; PPs, propelargonidins.

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inal liquid form. Commercial infant foods were purchased from local grocery stores and were freeze-dried before analysis. The concentration data are based on the original wet weight. Foods were extracted and purified according to a published method (9,10). All samples were analyzed in duplicate.

HPLC-MS/MS analyses. Analysis and quantification methods were described (9,10). Catechin glycoside and epicatechin glycoside, which were detected in some foods, were quantified as monomers. The quantification limits (signal-to-noise ratio = 10) of our method ranged from 0.076 to 0.191 ng (injection on column) for the individual oligomers and the polymers. The relative SD of the quantification was 4.2% for total PAs and 7.9 ± 3.4% for individual oligomers in a control blueberry sample ($n = 38$).

Estimation of PA intakes. The daily intake of PAs was estimated using data from the USDA's Continuing Survey of Food Intakes by Individuals (CSFII) of 1994–1996: mean quantities of common foods consumed per person per day (11). The mean daily intake of PAs by infants was estimated on the basis of the recommendations of the American Academy of Pediatrics (12), which were detailed as feeding guidelines by pediatricians for convenient use by parents (13).

RESULTS

The concentrations of PAs in 41 kinds of foods are listed in **Table 1**. All cultivars of apples and the sauce and juice derived directly from them are considered to be one kind. Grapes and wine are considered to be different kinds of food. There are also 57 foods containing no detectable PAs (see supplemental data).[5] Fruits were found to be the major source of PAs in the diet. In general, vegetables are not an important source of PAs. Of the 19 different vegetables tested, PAs were detected only in Indian squash. Minor cereals such as sorghum and barley contain PAs, whereas they are not detected in the staple crops such as corn, rice, and wheat. Wine, beer, and some commonly consumed fruit juices are good sources of PAs, whereas coffee is not.

The PAs in different foods vary considerably in terms of the total content and distribution of oligomers and polymers. A few foods, such as cashew nuts and black beans, contain only monomers and dimers, whereas most of the foods contain PAs with a wide range of DP values (1–10, >10). PAs with DP >10, which had escaped detection in early studies, were found to be the principal components (>50 g/100 g) in 21 kinds of foods in Table 1.

Different types of PAs were found in various foods. The homogeneous B-type PCs, which consist of catechin and/or epicatechin as constituent units, were detected as the exclusive PAs in 20 kinds of foods (Table 1). Another seven kinds of foods also contained exclusively PCs. However, a small fraction of PCs in those foods contained A-type linkages or gallic acid esters. In addition to the PCs, PPs and PDs were detected as minor components in various foods.

PA-containing ingredients have been used in complementary infant foods. PA concentrations in commercial infant foods were also determined (see supplemental data).[5] Because the consumption survey for complementary infant foods is not available, the mean intake of PAs by infants was estimated on the basis of the recommendations of the American Academy of Pediatrics (12). The estimations were also based on the results of our survey, which indicates that 25% of infant cereals, 90% of infant juices, and 85% of fruit-based infant foods contain PAs. According to the feeding guidelines (13), complementary infant feeding starts with infant cereals and juice for infants 4–6 mo of age with a recommended median serving amount of 12.5 g/d or 90 mL/d. Infant foods based on fruits are introduced after 6 mo. The recommended median serving amounts are 35 g of cereal, 125 mL of juice, and 100 g of cooked/mashed fruits per day. Assuming that there is no preference in the selection of the foods, the daily intake of PAs was estimated to be 1.3 mg/d for infants 4–6 mo old and 26.9 mg/d for infants 6-10 mo old.

The mean daily PA intakes of different age and gender groups in the United States were estimated and are shown in **Table 2**. The mean daily intake of PAs in the population of all ages (>2 y old) in the United States is estimated to be 53.6 mg/person excluding the monomers, or 57.7 mg/person including the monomers. Of the PAs ingested, ~73.9% have a DP >3. Raw apple is the major contributor to dietary PA intake (32.0%), followed by chocolate (17.9%) and grapes (17.8%). The PA intake in different age and gender groups varies. The 2–5 and 6–11 y old age groups and men >60 y old consumed more PAs than other groups because they included more fruits, especially apple, in their diet. The intake of PAs by adolescents was below the mean value.

DISCUSSION

In the present study, homogeneous B-type PCs isolated from cocoa or blueberry, which contain (+)-catechin and (–)-epicatechin as constituent units, were used as standards to quantify PAs in all foods. A majority of the foods in Table 1 contain PCs as exclusive components such as cocoa and blueberries. No biases in PA concentration due to the standards used are expected in these cases. However, PAs other than the homogeneous B-type PCs have been found in various foods. It was shown that PAs containing (epi)catechin gallic acid ester or (epi)gallocatechin as subunits had a lower fluorescent response compared with the PCs isolated from cocoa (8). Nevertheless, the biases on total PA contents should not be significant because (epi)catechin was found to be the predominant constituent unit in most foods containing PAs other than PCs (10).

PAs in foods cover a wide range of DP values. We presented the concentrations of monomers, dimers, and trimers separately because recent studies suggested that these low-molecular-weight PA oligomers (DP ≤ 3) could be absorbed intact in the gastrointestinal tract. Déprez et al. (14) demonstrated that (+)-catechin and PA dimers and trimers were permeable through the Caco-2 human intestinal cell line. The permeability of a PA polymer with a mean DP of 6 was ~10 times lower, suggesting that PA dimers and trimers could be absorbed in vivo and that polymers could not. Absorption of the dimers was confirmed by detection of PC B5 in human blood after the subjects consumed PC-rich chocolate (15).

We presented the concentration of all PAs with DP > 3 collectively in 3 groups because similarities in their absorption mode have been postulated. Unlike the lower oligomers, PAs with DP > 3 appear not to be absorbed directly from the gastrointestinal lumen (14) but are thought to depolymerize into mixtures of epicatechin monomer and dimers in the acidic environment of the stomach. The resultant monomers and dimers were absorbed in the small intestine (16). Our observation in pigs showed that depolymerization of ingested polymers (DP > 10) was not significant in the stomach 4 h after eating (Gu and Prior, unpublished data). It was suggested that the majority of PAs transit into the small intestine intact (17) and are degraded mainly by colonic microflora in the cecum and large intestine. Déprez et al. (18) reported that incubation of polymeric PCs with human colonic microflora in vitro in anoxic conditions completely degraded the PCs after 48 h. The degradation products included phenylacetic, phe-

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[5] A list of foods containing no detectable proanthocyanidins and concentrations of proanthocyanidins in infant foods is provided as supplemental data in the online posting of this paper at http://www.nutrition.org.

TABLE 1

Concentration of PAs in common foods[1,2]

No.	Food	Monomers	Dimers	Trimers	4–6 mers	7–10 mers	>10 mers	Total PAs	Moisture %	Type
					mg/100 g (fresh weight foods) mg/L (beverages)					
	Fruits									
1a	Blueberries, cultivated highbush	4.0 ± 1.5	7.2 ± 1.8	5.4 ± 1.2	19.6 ± 3.4	14.5 ± 2.0	129.0 ± 47.3	179.8 ± 50.8	85.0	PC
1b	Blueberries, lowbush	3.4 ± 0.5	9.0 ± 0.5	6.8 ± 0.4	25.7 ± 1.2	27.8 ± 1.3	260.4 ± 11.7	331.9 ± 14.0	85.0	PC
2	Cranberries	7.3 ± 1.5	25.9 ± 6.1	18.9 ± 3.4	70.3 ± 13.1	62.9 ± 14.7	233.5 ± 49.1	418.8 ± 75.3	87.2	A, PC
3	Blackberries	3.7 ± 2.2	6.7 ± 2.9	3.6 ± 1.9	7.3 ± 5.0	4.2 ± 4.5	1.5 ± 0.0	27.0 ± 17.5	86.9	PC
4	Marion berries	0.9 ± 0.0	3.4 ± 0.1	2.4 ± 0.0	2.2 ± 0.0	ND	ND	8.9 ± 0.1	86.9	PC
5	Choke berries	5.2 ± 0.2	12.5 ± 0.4	10.3 ± 0.3	40.3 ± 0.8	52.9 ± 3.1	542.6 ± 42.9	663.7 ± 47.7	71.8	PC
6	Raspberries	4.4 ± 3.4	11.5 ± 9.8	5.7 ± 5.5	7.7 ± 5.2	0.9 ± 2.2	ND	30.2 ± 23.4	85.8	PP, PC
7	Strawberries	4.2 ± 0.7	6.5 ± 1.3	6.5 ± 1.2	28.1 ± 6.5	23.9 ± 3.5	75.8 ± 13.4	145.0 ± 24.9	91.1	PP, PC
8	Blackcurrants	0.9 ± 0.2	2.9 ± 0.4	3.0 ± 0.3	10.6 ± 1.7	9.9 ± 1.4	122.4 ± 28.0	147.8 ± 33.0	79.4	PC, PD
9	Cherries	4.2 ± 1.1	2.8 ± 0.7	2.8 ± 0.5	6.5 ± 0.8	1.9 ± 0.1	ND	8.2 ± 03.2	80.2	PC
10a	Green grapes	1.0 ± 0.1	2.3 ± 0.0	1.9 ± 0.1	8.4 ± 0.3	9.2 ± 0.6	58.9 ± 14.2	81.5 ± 15.0	80.7	PC, PD
10b	Red grapes	0.8 ± 0.2	2.0 ± 0.3	1.5 ± 0.2	6.1 ± 0.9	6.2 ± 1.1	44.6 ± 9.9	61.0 ± 12.3	80.4	PC, PD
10c	Grape seed (dry)	660.3 ± 8.3	417.3 ± 4.8	290.2 ± 4.5	664.0 ± 8.2	400.3 ± 31.3	1100.1 ± 86.3	3532.3 ± 105.8		PC
11a	Apple, red delicious, with peel	9.6 ± 0.9	13.8 ± 0.6	9.3 ± 0.4	30.2 ± 1.2	25.4 ± 1.2	37.6 ± 2.6	125.8 ± 6.8	86.1	PC
11b	Apple, red delicious without peel	6.8 ± 0.9	11.3 ± 1.6	7.2 ± 1.0	24.3 ± 3.4	20.3 ± 3.1	28.7 ± 7.1	98.7 ± 17.0	86.7	PC
11c	Apple, golden delicious, with peel	4.7 ± 0.2	10.2 ± 0.2	6.3 ± 0.1	22.8 ± 0.6	19.5 ± 0.7	27.7 ± 2.9	91.1 ± 4.7	87.0	PC
11d	Apple, golden delicious, without peel	4.1 ± 0.1	9.4 ± 0.5	5.8 ± 0.5	21.2 ± 2.6	17.5 ± 3.2	22.4 ± 5.7	80.4 ± 12.4	86.9	PC
11e	Apple, granny smith	7.5 ± 1.0	15.0 ± 2.3	9.1 ± 1.5	32.9 ± 5.9	30.1 ± 6.1	46.3 ± 9.5	141.0 ± 26.1	85.7	PC
11f	Apple, gala	5.9 ± 0.4	9.5 ± 0.3	6.2 ± 0.2	21.3 ± 1.5	18.7 ± 1.4	30.7 ± 5.6	92.4 ± 8.4	86.0	PC
11g	Apple, fuji	6.5 ± 1.7	9.9 ± 2.6	6.1 ± 1.4	19.1 ± 4.3	13.8 ± 2.8	14.2 ± 3.1	69.6 ± 15.8	84.2	PC
11h	Apple sauce	2.3 ± 0.0	6.0 ± 0.0	3.0 ± 0.0	10.7 ± 0.1	8.3 ± 0.1	16.9 ± 0.4	47.2 ± 0.6	88.3	PC
12a	Peaches	4.7 ± 1.4	7.0 ± 2.2	5.0 ± 1.4	17.7 ± 5.5	10.9 ± 3.7	22.0 ± 7.7	67.3 ± 20.9	88.3	PC
12b	Peach, canned heavy syrup	0.6 ± 0.1	2.3 ± 0.6	ND	ND	ND	ND	2.9 ± 0.6	79.3	PC
13a	Pears, green cultivars	2.0 ± 0.3	2.7 ± 0.4	2.0 ± 0.3	6.0 ± 1.1	5.4 ± 1.4	24.2 ± 15.3	42.3 ± 18.6	83.4	PC
13b	Pears	2.7 ± 1.5	2.8 ± 1.3	2.3 ± 0.9	6.5 ± 1.9	4.6 ± 1.0	13.1 ± 11.3	31.9 ± 7.8	83.4	PC
14	Nectarines	1.9 ± 1.2	2.3 ± 1.2	1.7 ± 0.8	6.0 ± 3.0	3.6 ± 1.9	7.3 ± 6.5	22.8 ± 14.6	89.0	PC
15a	Black plums	6.8 ± 0.1	16.0 ± 0.4	14.9 ± 0.3	49.9 ± 0.1	34.9 ± 0.2	115.3 ± 2.0	237.9 ± 3.1	87.9	A, PC
15b	Plums, black diamond	9.9 ± 0.6	23.4 ± 1.6	22.8 ± 1.4	64.7 ± 4.3	41.2 ± 2.1	94.6 ± 8.7	256.6 ± 18.7	87.9	A, PC
15c	Plums	11.4 ± 3.4	31.5 ± 7.4	23.9 ± 5.1	58.0 ± 12.5	33.8 ± 11.9	57.3 ± 24.4	215.9 ± 50.7	87.4	A, PC
16	Apricots	2.8 ± 0.0	3.1 ± 0.0	1.9 ± 0.0	4.9 ± 0.1	2.2 ± 0.0	0.8 ± 0.2	15.6 ± 0.4	86.3	PC
17a	Kiwis, gold	1.1 ± 0.1	1.6 ± 0.1	1.2 ± 0.0	5.0 ± 0.1	5.0 ± 0.2	ND	13.9 ± 0.4	85.0	PC
17b	Kiwis	0.6 ± 0.5	0.8 ± 0.1	0.7 ± 0.0	1.3 ± 0.8	0.2 ± 0.0	ND	3.7 ± 1.6	83.9	PC
18	Avocados	1.0 ± 0.8	1.5 ± 0.8	1.4 ± 0.4	3.2 ± 0.8	0.4 ± 0.7	ND	7.4 ± 4.3	72.0	A, PC
19	Mangos	2.3 ± 0.1	1.8 ± 0.0	1.4 ± 0.0	7.2 ± 0.5	ND	ND	12.8 ± 0.5	81.7	PC
20	Dates, Deglet Noor (fresh)	ND	1.8 ± 0.5	3.0 ± 0.5	5.9 ± 0.7	ND	ND	10.7 ± 1.6	22.5	PC
21	Bananas	0.2 ± 0.0	0.7 ± 0.1	0.8 ± 0.1	2.3 ± 0.4	ND	ND	4.0 ± 0.6	73.5	PC
	Vegetable									
22	Indian squash, raw	1.6 ± 0.2	2.0 ± 0.2	1.5 ± 0.1	4.6 ± 0.4	3.2 ± 0.3	3.5 ± 0.4	16.4 ± 1.6	93.3	PC
	Cereals and beans									
23a	Sorghum, sumac bran	27.8 ± 1.2	78.2 ± 3.4	99.2 ± 7.7	585.5 ± 50.0	734.3 ± 69.3	2440.4 ± 271.0	3965.4 ± 402.5	9.2	PC
23b	Sorghum, sumac whole grain	18.0 ± 0.1	35.4 ± 0.4	45.6 ± 0.7	224.0 ± 9.2	289.2 ± 5.9	1307.3 ± 34.6	1919.5 ± 45.5	9.2	PC
23c	Sorghum, hi-tannin whole grain	0.9 ± 0.2	8.0 ± 1.1	10.3 ± 1.1	85.1 ± 14.4	150.0 ± 26.2	1533.3 ± 395.01	787.6 ± 438.1	9.2	PC
23d	Sorghum, hi-tannin whole grain extrudate	7.6 ± 0.0	23.5 ± 0.1	21.2 ± 0.0	80.6 ± 0.3	76.1 ± 0.1	238.3 ± 2.1	447.3 ± 1.8	9.2	PC
24a	Pinto beans, raw	14.8 ± 0.9	32.0 ± 2.6	28.3 ± 2.1	125.9 ± 9.2	135.6 ± 10.4	459.6 ± 34.2	796.3 ± 58.7	11.0	PP, PC
24b	Pinto beans, simmered 2 h	1.7 ± 0.0	4.4 ± 0.4	3.9 ± 0.3	10.5 ± 5.5	4.3 ± 3.9	1.4 ± 0.6	26.3 ± 12.8	11.0	PP, PC
25	Small red beans	10.6 ± 0.0	19.4 ± 0.8	18.1 ± 0.6	80.0 ± 2.7	75.7 ± 2.4	252.9 ± 0.8	456.6 ± 7.5	12.0	PP, PC
26	Red kidney beans	21.9 ± 0.2	26.4 ± 0.7	29.1 ± 0.7	117.7 ± 2.8	105.3 ± 2.2	263.4 ± 4.1	563.8 ± 10.4	12.0	PP, PC
27	Barley	11.0 ± 0.3	21.4 ± 1.1	14.6 ± 1.0	27.2 ± 0.6	ND	ND	74.2 ± 3.0	9.4	PC
28	Black eye peas	14.0 ± 4.4	6.0 ± 0.2	6.1 ± 0.1	7.3 ± 0.3	ND	ND	33.3 ± 4.2	12.0	PC
29	Black beans	2.9 ± 0.0	5.2 ± 0.1	ND	ND	ND	ND	8.1 ± 0.1	11.0	PC
	Nuts									
30	Hazelnuts	9.8 ± 1.6	12.5 ± 3.8	13.6 ± 3.9	67.7 ± 20.3	74.6 ± 21.9	322.4 ± 102.5	500.7 ± 152.0	5.3	PC, PD
31	Pecans	17.2 ± 2.5	42.1 ± 5.4	26.0 ± 2.0	101.4 ± 10.4	84.2 ± 12.9	223.0 ± 59.1	494.1 ± 86.2	3.5	PC, PD
32	Pistachios	10.9 ± 4.3	13.3 ± 1.8	10.5 ± 1.2	42.2 ± 5.2	37.9 ± 4.9	122.5 ± 37.1	237.3 ± 52.0	4.0	PC, PD
33	Almonds	7.8 ± 0.9	9.5 ± 1.6	8.8 ± 1.7	40.0 ± 8.5	37.7 ± 8.4	80.3 ± 28.1	184.0 ± 48.2	5.2	PP, PC
34	Walnuts	6.9 ± 3.4	5.6 ± 0.9	7.2 ± 1.2	22.1 ± 3.3	5.4 ± 0.8	20.0 ± 9.3	67.3 ± 14.7	4.1	PC
35	Peanuts, roasted	5.1 ± 1.0	4.1 ± 0.7	3.7 ± 0.5	2.8 ± 0.2	ND	ND	15.6 ± 2.3	2.0	A, PC
35	Peanut butter	2.0 ± 0.9	3.0 ± 0.7	8.1 ± 3.5	ND	ND	ND	13.2 ± 5.2	1.3	A, PC
36	Cashews	6.7 ± 2.9	2.0 ± 0.4	ND	ND	ND	ND	8.7 ± 3.2	5.2	PC

TABLE 1 *(continued)*

Concentration of PAs in common foods[1,2]

No.	Food	Monomers	Dimers	Trimers	4–6 mers	7–10 mers	>10 mers	Total PAs	Moisture %	Type
					mg/100 g (fresh weight foods) *mg/L* (beverages)					
	Beverages and snacks									
37	Baking chocolate, unsweetened	198.5 ± 3.0	206.5 ± 15.4	130.9 ± 15.0	332.6 ± 58.4	216.4 ± 56.0	551.0 ± 186.8	1635.9 ± 334.6	2.4	PC
37	Black chocolate	31.4 ± 0.2	31.2 ± 0.9	21.1 ± 0.8	55.5 ± 3.5	38.5 ± 3.0	68.2 ± 8.8	246.0 ± 0.3	1.3	PC
37	Milk chocolate	26.9 ± 3.0	26.2 ± 2.5	19.3 ± 2.6	51.4 ± 9.8	35.3 ± 7.2	32.8 ± 9.2	192.0 ± 28.8	1.3	PC
37	Chocolate milk	4 ± 1	22 ± 0	ND	ND	ND	ND	26 ± 2	82.3	PC
38	Red wine	20 ± 1	40 ± 1	27 ± 1	67 ± 2	50 ± 1	110 ± 2	313 ± 5	88.5	PC, PD
39	Beer	4 ± 0	11 ± 1	3 ± 0	4 ± 0	ND	ND	23 ± 2	95.2	PC, PD
2b	Cranberry juice cocktail	6 ± 0	29 ± 0	17 ± 0	49 ± 1	41 ± 1	89 ± 3	231 ± 2	85.5	A, PC
10d	Grape juice	18 ± 0	34 ± 0	19 ± 0	80 ± 0	69 ± 0	303 ± 2	524 ± 2	87.0	PC, PD
11i	Apple juice	1 ± 0	2 ± 0	1 ± 0	4 ± 0	1 ± 0		9 ± 0	87.9	PC
	Spices									
40	Cinnamon, ground	23.9 ± 1.3	256.3 ± 11.3	1252.2 ± 62.2	2608.6 ± 140.3	1458.3 ± 116.1	2508.8 ± 92.9	8108.2 ± 424.2	9.5	A, PP, PC
41	Curry powder	ND	9.5 ± 0.2	22.9 ± 0.5	41.8 ± 1.5	ND	ND	74.2 ± 2.2	9.5	A, PC

[1] Values are means ± SD, $n = 4–8$.

[2] Monomers, dimers, and trimers are listed separately. Tetramers through hexamers are pooled together as 4–6 mers. Polymers with DP > 10 are quantified collectively and listed as >10 mers. The moisture contents of the fresh fruits and the type of PAs are also presented, so that the PA contents can be converted to a dry weight basis.

Abbreviations and symbols: ND, not detected. The PP, PC, and PD are propelargonidins, procyanidins, and prodelphinidins, respectively. "A" indicates the existence of A-type PAs. The same number indicates the same kind of food, with different varieties within one kind of food labeled a, b, c, d, and so on.

nylpropionic, and phenylvaleric acids. These phenolic acids have been suggested to be the major metabolites of oligomeric and polymeric PAs in healthy humans (19).

Epidemiologic studies suggested an association between ingestion of polyphenols, especially flavonoids, and the prevention of diseases. Many authors have estimated the daily intake of flavonoids based on food composition and consumption survey data. Estimated data from several countries are largely consistent. The overall flavonoid (flavonols and flavones) intake in a population of women in the United States was estimated recently to be 24.6 ± 18.5 mg/d, with quercetin as the major contributor (70.2%) (4). The mean intake of flavonoid (including flavonols, flavones, and flavanones) was estimated to be 24.2 ± 26.7, 28.6 ± 12.3, and 25.9 mg/d in the populations of Finland, Denmark, and the Netherlands, respectively (5–7). However, these authors did not include monomeric, oligomeric, and polymeric flavan-3-ols in their estimation. Apples have been identified as a major dietary source of flavonoids in epidemiologic studies. The concentration of flavonoids other than PAs in fresh apples was determined to be 5.3 mg/100 g (3). The concentrations of PAs in various cultivars of apples were determined in our study to be in a range of 69.6–141.0 mg/100 g. Thus, the total flavonoid content in apples used in previous studies was significantly lower than the actual value. A similar situation may exist for other foods that contain PAs. As a result, underestimation of the total flavonoid intakes could be enormous.

Flavan-3-ol monomers are considered to be different from PAs by definition. Their ingestion was studied previously. Tea is known as a major source of flavan-3-ol monomers (and

TABLE 2

Estimation of the mean daily intake of PAs in the population of different age and gender groups in the United States

Age and gender group	Monomers	Dimers	Trimers	4–6 mers	7–10 mers	>10 mers	Total[1]
				mg/(d · person)			
Infants (4–6 mo)	0.2	0.4	0.3	0.4	0.0	0.0	1.3 (1.1)
Infants (6–10 mo)	2.3	4.8	3.0	8.5	5.4	3.0	26.9 (24.6)
2–5 y	3.9	5.9	4.4	13.4	10.7	30.0	68.2 (64.3)
6–11 y	4.4	6.2	4.6	13.6	10.6	25.8	65.1 (60.8)
12–19 y, male	4.3	5.7	4.1	11.1	8.0	17.7	50.9 (46.6)
12–19 y, female	3.5	4.8	3.5	10.5	7.9	19.7	49.9 (46.4)
20–39 y, male	4.9	8.2	5.0	12.5	8.3	18.3	57.4 (52.3)
20–39 y, female	3.5	5.3	3.7	10.7	7.8	18.3	49.4 (45.8)
40–59 y, male	5.0	8.2	5.4	14.3	9.8	21.7	64.6 (59.4)
40–59 y, female	4.0	6.0	4.3	12.6	9.2	20.6	56.7 (52.7)
>60 y, male	4.8	7.8	5.5	16.0	11.4	25.1	70.8 (66.0)
>60 y, female	3.4	5.4	4.0	12.3	9.0	20.6	54.6 (51.2)
Mean for >2 y	4.1	6.4	4.5	12.5	9.1	21.0	57.7 (53.6)

[1] The intakes of PAs excluding the monomers are shown in parentheses.

oxidative derivatives) but not an important source of PAs, because few PAs have been detected in green leaves (20,21). Arts et al. (22) estimated that the mean intake of flavan-3-ol monomers in the Netherlands was 50 mg/d, with tea as the major contributor (65.2–87.3%) followed by chocolate and apple. He also pointed out that the mean daily intake of flavan-3-ol monomers in the United States should be lower than that in Denmark. The daily intakes of flavan-3-ol monomers from tea were estimated to be in the range of 12.7–34.2 mg/person for adults in the United States based on the data of Lakenbrink et al. (20). The total daily intake of flavan-3-ol monomers is estimated to be 17.1–38.6 mg/person for adults in the United States after flavan-3-ols from other foods (Table 1) are included. The mean daily intake of oligomeric and polymeric PAs (53.6 mg/person, Table 2) is higher than that of monomeric flavan-3-ols, and is twice as high as the combined overall intake of other flavonoids, which include flavonols, flavones, and flavanones. Based on these discussions, we conclude that PAs are the major flavonoids ingested in Western diets.

Complementary infant foods are necessary to meet increasing nutritional needs and for weaning. The mean daily intake of PAs for infants 6–10 mo old is estimated to be 3.1 mg/kg body weight, which is four times higher than the mean daily intake in adults of >20 y old (0.77 mg/kg body weight). Information concerning the influence of such high intake of PAs or other phytochemicals on the health and growth of infants is scarce. Increasing evidence suggests that exposure to these nutrients in early infancy may have long-term effects in later life (23).

In conclusion, this study demonstrates that many foods contain substantial amounts of PAs. PAs in these foods are different in terms of concentration, distribution of oligomers and polymers, constituent flavan-3-ol units, and interflavan linkages. PAs account for a major fraction of the total flavonoids ingested in Western diets. PAs should be taken into account when studying the epidemiologic association between flavonoid intake and chronic diseases. This study provided the first opportunity to examine this association.

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