Catechin Contents of Foods Commonly Consumed in The Netherlands. 1. Fruits, Vegetables, Staple Foods, and Processed Foods

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Catechins, compounds that belong to the flavonoid class, are potentially beneficial to human health. To enable epidemiological evaluation of these compounds, data on their contents in foods are required. HPLC with UV and fluorescence detection was used to determine the levels of (+)-catechin, (−)-epicatechin, (+)-gallocatechin (GC), (−)-epigallocatechin (EGC), (−)-epicatechin gallate (ECg), and (−)-epigallocatechin gallate (EGCg) in 24 types of fruits, 27 types of vegetables and legumes, some staple foods, and processed foods commonly consumed in The Netherlands. Most fruits, chocolate, and some legumes contained catechins. Levels varied to a large extent: from 4.5 mg/kg in kiwi fruit to 610 mg/kg in black chocolate. (+)-Catechin and (−)-epicatechin were the predominant catechins; GC, EGC, and ECg were detected in some foods, but none of the foods contained EGCg. The data reported here provide a base for the epidemiological evaluation of the effect of catechins on the risk for chronic diseases.

Keywords: *Catechins; flavanols; flavonoids; fruits; vegetables; legumes; chocolate*

INTRODUCTION

Observational studies show that fruits and vegetables protect against cancer and cardiovascular diseases (Law and Morris, 1998; Ness and Powles, 1997; Steinmetz and Potter, 1996). An important question is which compounds might be responsible for this protective effect. Flavonoids, secondary plant metabolites with antioxidant activity, are potentially protective compounds. The effects of flavonoids on the cancer process, the immune system, and hemostasis have been reported in cell systems and animals (Middleton and Kandaswami, 1994). The average total intake of flavonoids in the United States was estimated to be 1 g/day. Catechins (Figure 1), one of the six classes of flavonoids, contributed one-fifth of the total estimated intake (Kuhnau, 1976). Catechins are the principal components of tea; they make up 3−10% of black tea solids and 30−42% of green tea solids (Balentine et al., 1997). Epidemiological studies have shown that tea may reduce the risk for certain cancers (Kohlmeier et al., 1997; Blot et al., 1996), coronary heart disease, and stroke (Tijburg et al., 1997). However, the protective effect of tea is not undisputed. Catechins are also present in fruits and vegetables, and this may partially account for the ambiguity in the epidemiological data on tea. Still, quantitative data on catechin contents of foods are, as yet, largely absent, incomplete, or unreliable.

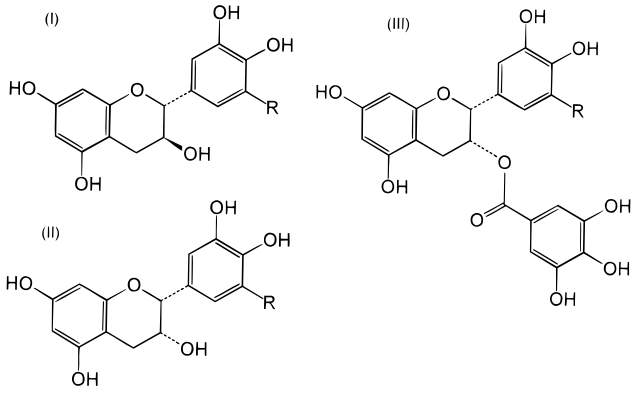

Figure 1. Chemical structures of catechins: (**I**) R = H (+)-catechin, R = OH (+)-gallocatechin (GC); (**II**) R = H (−)-epicatechin, R = OH (−)-epigallocatechin (EGC); (**III**) R = H (−)-epicatechin gallate (ECg), R = OH (−)-epigallocatechin gallate (EGCg).

Herrmann and co-workers started in the 1970s with the analyses of catechins in fruits using thin-layer chromatography methods with spectrophotometric measurements (Berger and Herrmann, 1971; Mosel and Herrmann, 1974b; Stöhr and Herrmann, 1975a,b). These methods have been used often since. Their major drawback is that they respond not only to catechins but also to other compounds, which may result in an overestimation of the catechin content (Solich et al., 1996; Sarkar and Howarth, 1976). Over the past 10 years, analytical methods have evolved and nowadays selective and sensitive HPLC methods are available. However, reported data on catechin contents of foods are incomplete and mostly limited to (+)-catechin and (−)-epicatechin. Other catechins may be important to human health as well. For example (−)-epigallocatechin

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gallate (EGCg) (Figure 1) has been suggested as an important compound in the prevention of cancer (Jankun et al., 1997).

Epidemiological research requires reliable and representative data on catechins in foods. This means that samples from different sale outlets, seasons, and years should be included to ensure that natural variation is taken into account. The goal of the present study was to provide these data for commonly consumed foods. In this paper we present data on the catechin [(+)-catechin, (−)-epicatechin, (+)-gallocatechin (GC), (−)-epigallocatechin (EGC), (−)-epicatechin gallate (ECg), and (−)-epigallocatechin gallate (EGCg)] contents of 24 types of fruits, 27 types of vegetables and legumes, some staple foods, and a number of processed foods.

EXPERIMENTAL PROCEDURES

Sample Collection and Preparation. Fruits and vegetables were selected using data from the Dutch National Food Consumption Survey 1992 and literature data regarding catechin-containing foods. Auction supply data were used to select the common varieties of apples and pears. The Dutch National Food Consumption Survey 1992 consists of a nationwide 2-day dietary record among 6218 males and females aged 1−92 years. Fruits consumed in quantities in excess of 1 g/person/day and vegetables consumed in excess of 3 g/person/day were selected. These limits were chosen because products eaten in small quantities, but with high levels of catechins, may make substantial contributions to the intake. We sampled 24 types of fruits, 27 types of vegetables and legumes, some staple foods (rice, macaroni, and wholemeal bread), and a number of processed foods (chocolate, jam, raisins, and currants). Seven varieties of apples and four varieties of pears were included because these fruits are popular in The Netherlands and supposedly contain relatively high levels of catechins. Rice and macaroni were analyzed after preparation according to the instructions on the packing. Canned and jarred products were sampled if they contributed substantially to the total consumption of a certain food in The Netherlands.

Fresh fruits and vegetables were sampled four times: in August 1997, December 1997, April 1998, and August 1998. We started in April 1997 with apples and pears only, to test the sampling and processing procedures. Fresh products were bought on the same day at three locations: an outlet of a nationwide supermarket chain (Albert Heijn), a local grocery, and an open-air street market. A minimum of 1 kg (or 0.5 kg for very small foods such as berries) or at least three units was sampled at each location. After removal of the nonedible parts, these samples were combined per product to a composite in proportions reflecting sales at the three locations. On average, the supermarket accounted for 64%, the grocery for 18%, and the street market for 18% of the sample. Three major brands of each canned or jarred product were bought in the supermarket in April 1998. The contents were allowed to drain for 5 min in a household colander, and the brands were mixed in equal proportions. All composites were either chopped under liquid nitrogen or cut into smaller pieces and frozen in liquid nitrogen and stored at −20 °C until freeze-drying was started within 2 weeks. After freeze-drying, the samples were ground to a powder and stored at −20 °C.

Analytical Methods. Six major catechins [(+)-catechin, (−)-epicatechin, GC, EGC, ECg, and EGCg] (Figure 1) were determined using a method described previously in detail (Arts and Hollman, 1998). In short, catechins were extracted from freeze-dried foods with aqueous methanol at room temperature for 1 h in a mechanical shaker. The concentration of methanol was either 70 or 90%, depending on the previously determined optimal concentration for the product. Sample extracts were injected into a gradient reversed phase HPLC-UV-fluorescence system, operated at 30 °C. (+)-Catechin and (−)-epicatechin were measured by means of fluorescence (280 nm excitation, 310 nm emission wavelengths); the other catechins were measured by means of UV (270 nm).

Table 1. Detection Limits of Catechins

	fluorescence		UV	
	standard (μg/mL)	apple[a] (mg/kg of fresh wt)	standard (μg/mL)	apple[a] (mg/kg of fresh wt)
(+)-catechin	0.008	0.1	0.13	1.8
(−)-epicatechin	0.007	0.1	0.09	1.2
EGC			0.29	3.9
EGCg			0.03	0.4
ECg			0.03	0.4

[a] Detection limits in apple are calculated from the standard data as follows: [value in μg/mL)/sample weight] × 50 mL × (fraction dry weight); sample weight is 0.5 g, fraction dry weight is 0.136

Chocolate was not freeze-dried before extraction, but frozen and ground to a fine powder using a handmill (model 1101, Alexander Werk, Germany). Extraction at room temperature was inefficient, probably due to the high content of hard fat in this product. Reflux extraction (90 °C) of 0.5 g of homogenized chocolate in 25 mL of 90% methanol in water for 30 min under a continuous nitrogen flow improved the catechin yield by ∼40%. Longer extraction (up to 90 min) did not further enhance yield.

In addition to the acetonitrile/phosphate buffer gradient described in the paper by Arts and Hollman (1998), a second gradient using methanol/phosphate buffer was developed. The solvents used for separation were 5% methanol (eluent A) and 40% methanol (eluent B) in phosphate buffer (0.025 M, pH 2.4). The gradient was as follows: 0−30 min, linear gradient from 5 to 90% B; 30−37 min, isocratic at 90% B; 37−40 min, linear return to 5% B; 40−50 min, isocratic at 5% B to re-equilibrate. This gradient was used to obtain baseline peak separation for some samples, and it was used for additional peak identification.

Analytical Quality Control. Because of the large variety of products analyzed and the extended series of analysis, quality control and confirmation of the identity of the peaks were considered to be important issues. Control samples consisting of either apple or black grape were included at the beginning and end of each series of analysis. The within- and between-run reproducibilities of the analysis of these samples were reported previously (Arts and Hollman, 1998) and considered to be satisfactory (<10%). The catechin content of the control sample was recorded after each series of analysis and had to be within the confidence limits (mean ± 2 SD). The recovery of catechins was determined by spiking apple with a known amount of standard compound; recoveries ranged from 92 to 105% (Arts and Hollman, 1998). Limits of detection are shown in Table 1. All samples were analyzed in duplicate.

The peaks of interest were identified by two methods. First, all samples were separated with both the acetonitrile and the methanol gradient described under Analytical Methods. The retention times of the peaks were compared to the retention times of pure standards. Peak identity was confirmed when peak retention times were identical to those of the pure standards in both mobile phases. Second, all samples were analyzed with diode array detection (HP 1040A upgraded version, Hewlett-Packard, Palo Alto, CA), and the peak spectra were compared to spectra of the pure compounds. Peak purity was also determined by diode array detection. For all samples peak purity was satisfactory with at least one of the mobile phases; catechin contents were calculated on the basis of the chromatogram with the best peak purity.

Catechins in anthocyanin-rich fruits, such as red currants, cherries, and blackberries, were difficult to quantify by UV detection, because a large "hump" appeared in the chromatogram area where EGCg and ECg normally elute. The determination of (+)-catechin and (−)-epicatechin was not hindered because anthocyanins do not fluoresce at the chosen wavelengths. We developed a method to remove the anthocyanins from the sample, without substantial loss of catechins. Twenty-six milliliters of a water/ethyl acetate (4:96 v/v) solution was

added to 1 g of freeze-dried sample. The extract was placed in a mechanical shaker for 60 min (250 rpm) at room temperature. After extraction, 2 mL of the clear supernatant was evaporated to dryness in a heating module (Pierce, Rockford, IL) at 50 °C. The residue was redissolved in 90% aqueous methanol, passed through a 0.45 μm Acrodisc filter, and injected. Recoveries were determined by adding known amounts of catechins immediately after the addition of ethyl acetate. Recoveries ranged from 90% for (−)-epicatechin to 114% for EGC. All anthocyanin-rich fruits were analyzed using this extraction method, but none of them contained EGCg or ECg. Thus, all data presented under Results are based on simple aqueous methanol extraction.

RESULTS AND DISCUSSION

Catechins were found particularly in fruits, but black chocolate contained the highest levels of catechins. Vegetables and legumes were poor dietary sources of catechins: only rhubarb, broad beans, and marrowfat peas contained catechins (Table 2). The studied staple foods did not contain catechins. Foods with an average total catechin content >100 mg/kg of fresh edible weight were dark chocolate (459.8−610.0 mg/kg), milk chocolate (153.0−163.2 mg/kg), raw (493.7 mg/kg) and home-cooked (206.3 mg/kg) broad beans, black grapes (203.9 mg/kg), blackberries (187.4 mg/kg), cherries (117.1 mg/kg), apricots (110.1 mg/kg), and some apple varieties (71.1−115.4 mg/kg of fresh edible weight). (−)-Epicatechin is the quantitatively most important catechin. EGCg was not detected in any of the foods, whereas GC, EGC, and ECg were limited to a few products.

Literature data on catechin contents of foods determined with appropriate methods (HPLC separation, proper peak identification, analysis of edible parts only, and data reported on a fresh weight basis) are summarized in Table 3. The remaining data reported in the literature are either qualitative only, quantitative but based on less specific separation (thin-layer chromatography, TLC) or detection methods (e.g., vanillin−HCl or Folin−Ciocalteu), or reported on a dry weight basis. Although these data have been obtained with methods now considered to be obsolete, we will compare them to our data whenever no other data are available. Data obtained with vanillin−HCl or Folin−Ciocalteu reagents without chromatographic separation, for which results are expressed as "total catechin equivalents", will not be used.

Apples and Pears. Our data for apples correspond well with most of the data reported in the literature (Table 3). Varietal differences were relatively small (Table 2). Escarpa and González (1998) found exceptionally high levels of (+)-catechin in four varieties of apple (among which were Golden Delicious and Granny Smith). The (−)-epicatechin levels reported by them are in the range usually found, and their analytical method seems to be adequate. It is not clear what may have caused the difference between their data and other reported data. Possibly, outlying extreme samples were included in the limited number of samples taken: values for six individual apples are given, instead of a composite of several apples. In our study, the reported catechin content of each apple variety is based on the analysis of 12 samples of 1 kg purchased at 3 locations in 4 periods.

Two previous studies on the catechin contents of various pear cultivars reported data similar to ours (Table 3). Both (+)-catechin and (−)-epicatechin are present in most pear cultivars, but (+)-catechin levels are sometimes very low. In general, pears contain lower catechin levels than apples.

Grapes. Our data for black grapes are considerably higher than those reported by Oszmianski and Lee (1990) for red grapes; they did not report on ECg (Table 3). Removal of the seeds, which was done in their study, but not in ours, may have contributed to the difference. However, they themselves report that their data on red grapes are relatively low, even compared to white grapes grown in the same area. Qualitative data confirm the presence of (+)-catechin, (−)-epicatechin, and ECg in black and white grape seeds and skins (Santos-Buelga et al., 1995; Escribano-Bailón et al., 1995; Oszmianski and Sapis, 1989). However, ECg was not present in the skins of all varieties that were tested (Escribano-Bailón et al., 1995). Similarly, white grapes contained ECg in only one of our four sampling periods, thus pointing to a varietal effect.

Other Fruits. Risch and Herrmann (1988) detected (−)-epicatechin in peach, whereas we did not (Table 3). Because their levels for (+)-catechin are substantially higher as well, this may have been due to varietal differences. (−)-Epicatechin was, for example, also absent from Desert Gold peaches, a variety that Risch and Herrmann did not study (Van Gorsel et al., 1992). The variety of the peaches we sampled has not been recorded.

Van Gorsel (1992) reported the presence of (+)-catechin and (−)-epicatechin in nectarines, whereas we found only (+)-catechin. Although van Gorsel used appropriate separation and detection methods, peak identity was confirmed only by comparing retention times with those of known standards. This can easily lead to the erroneous reporting of a compound.

We found low levels of (−)-epicatechin in kiwi fruit. Dawes and Keen (1999) reported similar levels in experimentally prepared kiwi fruit juice using HPLC methods, but they also detected trace amounts of (+)-catechin. Again, the reported difference in types of catechins present may be due to varietal differences.

We detected (+)-catechin and (−)-epicatechin in raspberry, which confirmed qualitative data by Rommel and Wrolstad (1993).

To our knowledge, no more studies have been published that employed HPLC with on-line detection to determine catechins in fruits. However, with colorimetric detection after TLC separation, Herrmann and co-workers (Hanefeld and Herrmann, 1976; Stöhr and Herrmann, 1975a,b) reported the presence of catechins in strawberries, black, white, and red currants, gooseberries, blueberries, and rhubarb. Quantitatively, our data are in the same order of magnitude. Herrmann and co-workers, however, occasionally report the presence of a certain catechin, for example, (−)-epicatechin in strawberry, which we did not detect. This is most likely due to their unspecific methods and lack of peak identity confirmation, although of course differences in variety may have contributed as well. Our data on strawberries confirm those reported by Van Gorsel et al. (1992), who did not detect (−)-epicatechin in juice made from strawberries either. Finally, our data confirm an early report on the presence of low levels of (−)-epicatechin in avocado (Ramírez-Martínez and Luh, 1973).

Legumes. In faba beans (*Vicia faba* L.), which belong to the same family as broad beans, (−)-epicatechin and EGC were detected (Helsper et al., 1993). We found (+)-catechin in addition to these compounds in broad beans,

Table 2. (+)-Catechin, (−)-Epicatechin, GC, EGC, and ECg in Raw and Processed Foods[a]

product	scientific name or brand name	catechin content (mg/kg of fresh edible wt)[b] (+)-catechin	(−)-epicatechin	others	total catechins
apple with skin	*Malus pumila* Mill.				
Cox's Orange Pippin		12.8 ± 1.67	96.2 ± 18.37	ND[c]	109.0
Elstar		12.4 ± 2.11	81.7 ± 12.41	ND	94.0
Golden Delicious		5.3 ± 0.70	74.2 ± 7.44	ND	79.5
Goudreinette		12.2 ± 0.89	103.2 ± 17.72	ND	115.4
Granny Smith		15.6 ± 4.33	74.8 ± 15.82	ND	90.3
Jonagold		4.0 ± 0.29	67.1 ± 7.26	ND	71.1
Jonagored		4.2 ±1.63	72.3 ± 22.33	ND	76.5
apple without skin	*Malus pumila* Mill.				
Cox's Orange Pippin		12.8	66.5	ND	79.3
Elstar		11.0 ± 3.29	66.4 ± 3.86	ND	77.4
Golden Delicious		4.4 ± 0.26	50.7 ± 8.54	ND	55.1
Goudreinette		9.6 ± 0.66	86.1 ± 14.81	ND	95.6
Granny Smith		16.5 ± 5.19	65.3 ± 15.24	ND	81.1
Jonagold		2.8 ± 0.57	48.3 ± 6.84	ND	51.2
Jonagored		3.0 ± 1.56	51.6 ± 17.17	ND	54.6
applesauce		6.9	54.1	ND	61.0
apricot	*Prunus armeniaca* L.	49.5 ± 43.68	60.6 ± 78.49	ND	110.1
avocado	*Persea americana* Mill.	ND	5.6 ± 2.91	ND	5.6
blackberry	*Rubus* sp.	6.6 ± 0.58	180.8 ± 21.39	ND	187.4
blueberry	*Vaccinium corymbosum* L.	ND	11.1 ± 1.00	ND	11.1
broad bean, raw	*Vicia faba* L.	128.3 ± 160.60	225.1 ± 184.78	EGC: 140.3 ± 127.90	493.7
prepared		81.6 ± 36.44	78.2 ± 40.93	EGC: 46.5 ± 23.17	206.3
canned		ND	ND	ND	ND
cherry, sweet	*Prunus avium* L.	21.7 ± 9.18	95.3 ± 24.84	ND	117.1
canned		ND	43.1	ND	43.1
cranberry	*Vaccinium oxycoccus* L.	ND	42.0	ND	42.0
currant, black	*Ribes nigrum* L.	7.0	4.7	ND	11.7
currant, white	*Ribes pallidum*	3.0	ND	7.0	10.0
currant, red	*Ribes rubrum-hybriden*	12.2 ± 4.35	ND	GC: 12.2 ± 10.85	24.4
gooseberry	*Ribes uva-crispa* L.	16.7 ± 3.63	ND	GC: 4.4 ± 6.27	21.2
grape, black	*Vitis vinifera* L.	89.4 ± 91.80	86.4 ± 71.20	ECg: 28.1 ± 37.93	203.9
white		24.7 ± 10.59	10.2 ± 5.18	ECg: 4.3 ± 8.54	39.2
kidney-bean, canned	*Phaseolus vulgaris* L.	16.6	3.5	ND	20.1
kiwi fruit	*Actinidia chinensis* Planch	ND	4.5 ± 1.05	ND	4.5
mango	*Mangifera indica* L.	17.2 ± 15.72	ND	ND	17.2
marrowfat pea, canned	*Pisum sativum* L.	ND	ND	GC: 43.3 EGC: 56.4	99.7
nectarine	*Prunus persica* Batsch	27.5 ± 2.42	ND	ND	27.5
peach	*Prunus persica* Batsch	23.3 ± 5.66	ND	ND	23.3
canned		18.7	ND	ND	18.7
pear with skin	*Pyrus communis* L.				
Conference		1.1 ± 0.36	29.5 ± 1.69	ND	30.6
Doyenne du Comice		0.6 ± 0.44	34.8 ± 16.34	ND	35.4
Guyot		1.9	37.0	ND	38.9
cooking pear[d]		9.6 ± 4.77	75.4 ± 32.42	ND	85.0
canned		1.8	2.6	ND	4.4
pear without skin	*Pyrus communis* L.				
Conference		0.4 ± 0.18	8.2 ± 2.72	ND	8.5
Doyenne du Comice		0.1	14.4	ND	14.6
cooking pear[d]		3.6 ± 0.18	29.6 ± 3.25	ND	33.3
prepared		3.3	21.2	ND	24.5
plum	*Prunus domestica* L.	33.5 ± 9.13	28.4 ± 31.89	ND	61.9
raspberry	*Rubus idaeus* L.	9.7 ± 2.57	82.6 ± 13.06	ND	92.3
rhubarb	*Rheum rhabarbarum* L.	21.7 ± 11.39	5.1 ± 3.30	ECg: 6.0 ± 3.71	32.8
- prepared		14.8 ± 10.61	3.8 ± 1.31	ECg: 4.9 ± 6.06	23.5
strawberry	*Fragaria ananassa* Duch.	44.7 ± 13.80	ND	ND	44.7
chocolate, black	Albert Heijn	132.4	327.4	ND	459.8
	Verkade	107.5	502.5	ND	610.0
chocolate, milk	Albert Heijn	38.3	124.9	ND	163.2
	Verkade	26.9	126.1	ND	153.0
chocolate candy bar	Mars	21.7	62.5	ND	84.2
currant		ND	ND	ND	ND
jam, apricot	Albert Heijn	4.7	5.0	ND	9.7
jam, cherry	Albert Heijn	1.6	9.0	ND	10.6
jam, forest fruit	Albert Heijn	0.7	15.7	ND	16.4
jam, strawberry	Albert Heijn	9.0	ND	ND	9.0
raisins		29.7	7.1	ND	36.8

[a] Data include seasonal and regional variation. EGCg was not detected in any food; no catechins were detected in banana (*Musa sapientum* L.), broccoli (*Brassica oleracea* L. cv. *botrytis L.*), Brussels sprout (*B. oleracea* L. cv. *gemmifera DC.*), carrot (*Daucus carota* L.), cauliflower (*B. oleracea* L. cv. *botrytis L.*), chicory (*Cichorium intybus* L.), Chinese cabbage (*B. oleracea* L. cv. *conica DC.*), cucumber (*Cucumis sativus* L.), endive (*Ci. endivia* L.), French bean (*Phaseolus vulgaris* L.), green olive (*Olea europaea* L.), green pea (canned) (*Pisum sativum* L.), leek (*Allium porrum* L.), lettuce (*Lactuca sativa* L. cv. *capitata L.*), macaroni, maize (*Zea mays* L. cv. *saccharata*), melon (*Cu. melo* L.), mushroom (*Agaricus campester* Fr.), onion (*A. cepa* L.), orange (*Citrus sinensis* L.), pineapple (*Ananas comosus* L. Merr.), potato (*Solanum tuberosum* L.), red beet (*Beta vulgaris* L. cv. *rubra* L.), red cabbage (*B. oleracea* L. cv. *rubra DC.*), rice (*Oryza sativa* L.), slicing bean (*Ph. vulgaris* L.), sweet red pepper (*Capsicum annuum* L.), sugar peas (*Pi. sativum* L. cv. *saccharatum*), tomato (*Lycopersicon esculentum* Mill.), white bean in tomato sauce (*Ph. vulgaris* L.), and wholemeal bread. [b] Annual mean ± SD, based on duplicate analyses for each season; the food was bought in only one season if no SD is given. [c] ND, not detected (see Table 1 for limits of detection). [d] Different varieties.

Table 3. Reported Data on Catechins in Foods, Determined by HPLC and Reported on a Fresh Weight Basis

product	content (mg/kg of fresh edible wt) (+)-catechin	(−)-epicatechin	other	remarks	reference
apple	trace−17	2−101	−[a]	16 varieties	Risch and Herrmann, 1988
apple without skin	−	10−140	−	3 varieties	Burda et al., 1990
apple without skin	0.2−55	14−81	−	5 varieties	Perez-Ilzarbe et al., 1991
apple without skin	28−182	19−111	−	4 varieties	Escarpa and Gonzalez, 1998
apricot	26−57	67−171	−	3 varieties	Risch and Herrmann, 1988
peach	50−129	3−15	−	4 varieties	Risch and Herrmann, 1988
pear	trace−10	5−59	−	10 varieties	Risch and Herrmann, 1988
pear	ND[b]−5	6−87	−	7 varieties	Amiot et al., 1995
plum	5−36	0−16	−	8 varieties	Risch and Herrmann, 1988
red grapes	<10	17.5−21.4	−	2 varieties, seeds removed	Oszmianski and Lee, 1990
sweet cherry	5−12	14−49	−	5 varieties	Risch and Herrmann, 1988

[a] −, not determined. [b] ND, not detected.

but it is not clear whether Helsper and co-workers determined (+)-catechin. All other available data on catechins in legumes are expressed as "total catechin equivalents" and are therefore unreliable estimates of catechin contents. A general trend of increasing "total catechin equivalent" content with increasing darkness of the legumes within one family can be observed (Moneam, 1990; Deshpande and Cheryan, 1987). We found that within the *Phaseolus* genus the total catechin content of the dark kidney bean was higher than the catechin content of the green beans.

Chocolate. We found very high levels of (+)-catechin (107.5−132.4 mg/kg) and (−)-epicatechin (327.4−502.5 mg/kg) in black chocolate (54% cacao) and lower but still substantial levels in milk chocolate (34% cacao). Previous research has shown the presence of (+)-catechin and (−)-epicatechin in cacao liquor (a major ingredient of chocolate) (Sanbongi et al., 1998) and in fresh and fermented cacao beans (Porter et al., 1991; Kim and Keeney, 1984). Fresh beans contained up to 43270 mg of (−)-epicatechin kg^{-1} of defatted sample (Kim and Keeney, 1984). On fermentation and roasting of cacao beans, the characteristic red-brown color of chocolate is formed, probably as a result of polymerization of catechins (Porter et al., 1991). Fermented cacao beans therefore contain ~90% lower catechin levels than fresh beans (Porter et al., 1991; Kim and Keeney, 1984). It is now determined that chocolate still contains significant amounts of (+)-catechin and (−)-epicatechin.

Seasonal Variation and Maturity. The seasonal variation was considerable for some products (e.g., black grapes, broad beans) and surprisingly small for others (e.g., apples). Factors that may have contributed to the seasonal variation are varietal differences, maturity of the product, and storage period. Part of the seasonal variation is undoubtedly due to differential availability of varieties over the year. Except for apples and pears, we did not sample specific fruit varieties, but purchased those that were available in the shops. Because all purchases were done at three locations, the analyzed samples are often mixtures of several varieties. Specific varieties were purchased only for apples and pears, and for these fruits the seasonal variation was relatively small. Part of the variation may have been caused by differences in maturity. For apples and pears, it has been shown that during growth there is a rapid decrease in catechin levels (Amiot et al., 1995; Burda et al., 1990). Data from the 1970s using TLC had shown similar rapid decreases preceding maturity for cherries (Stöhr et al., 1975) and strawberries (Stöhr and Herrmann, 1975a), but much less so for plums (Stöhr et al., 1975), and black currants (Stöhr and Herrmann, 1975b). However, no unripe fruits are generally for sale for consumers, and fluctuations are reportedly low during maturation in apples and pears (Amiot et al., 1995; Burda et al., 1990). Information on changes in catechins during maturation for fruits other than apples and pears is not available and may differ substantially. Although we took care to purchase ripe fruits only, small differences may have remained. Storage of apples, harvested at commercial maturity, under normal cold-storage conditions for over 6 months changed their (+)-catechin and (−)-epicatechin content only very marginal (Burda et al., 1990; Mosel and Herrmann, 1974a).

Home Preparation. Processing resulted in a decrease of the catechin content of foods, although the magnitudes of the decrease differed considerably. Rhubarb, broad beans, and cooking pears were analyzed both raw and prepared according to standard recipes. Total catechins in the prepared products were, respectively, 28, 58, and 26% lower than in the raw products. In general, it can be concluded that processing is not an important issue for catechins because they are present mainly in fruits, which are usually consumed raw. In contrast to flavonols in apple, which are almost completely found in the peel (Burda et al., 1990; Escarpa and Gonzalez, 1998), only a small part of the catechins was present in apple peel. Removing the apple peel resulted on average in a 23% decrease of the total catechin content. For pears this percentage was higher (45%).

Summary. Most fruits, some legumes, and chocolate contained catechins, whereas vegetables and staple foods did not. Amounts varied from low (4.5 mg/kg in kiwi fruit) to very high (610 mg/kg in black chocolate). Preparation of foods caused a decrease in catechin content, but because most catechin-containing foods are consumed raw, this is not likely to be an important determinant of catechin intake. (+)-Catechin and (−)-epicatechin were the most common catechins in foods. EGCg was not found, and GC, EGC, and ECg were found only occasionally. The data reported here provide a base for the epidemiological evaluation of the effect of catechins on the risk for chronic diseases.

ABBREVIATIONS USED

GC, (+)-gallocatechin; EGC, (−)-epigallocatechin; EGCg, (−)-epigallocatechin gallate; ECg, (−)-epicatechin gallate.

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