Catechin Contents of Foods Commonly Consumed in The Netherlands. 2. Tea, Wine, Fruit Juices, and Chocolate Milk

Ilja C. W. Arts,†,‡ Betty van de Putte,† and Peter C. H. Hollman*,†

State Institute for Quality Control of Agricultural Products (RIKILT), Wageningen, The Netherlands, and Department of Chronic Diseases Epidemiology, National Institute of Public Health and the Environment (RIVM), Bilthoven, The Netherlands

Catechins, compounds that belong to the flavonoid class, are potentially beneficial to human health. To enable an epidemiological evaluation of catechins, 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 8 types of black tea, 18 types of red and white wines, apple juice, grape juice, iced tea, beer, chocolate milk, and coffee. Tea infusions contained high levels of catechins (102−418 mg of total catechins/L), and tea was the only beverage that contained GC, EGC, ECg, and EGCg in addition to (+)-catechin and (−)-epicatechin. Catechin concentrations were still substantial in red wine (27−96 mg/L), but low to negligible amounts were found in white wine, commercially available fruit juices, iced tea, and chocolate milk. Catechins were absent from beer and coffee. 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; beverages; tea; wine; fruit juice; chocolate milk*

INTRODUCTION

A number of epidemiological studies have demonstrated a protective effect of tea against the development of certain cancers (Blot et al., 1996; Kohlmeier et al., 1997), coronary heart disease, and stroke (Tijburg et al., 1997). Catechins, or flavanols, one of the six classes of flavonoids, are the principal components of tea and may be responsible for its alleged protective effect. In vitro and in vivo experiments have shown that catechins are potentially beneficial to human health: they are strong antioxidants, anticarcinogens, anti-inflammatory agents, and inhibitors of platelet aggregation (Cook and Samman, 1996; Katiyar and Mukhtar, 1996; Kohlmeier et al., 1997). Not all epidemiological studies, however, have found a protective effect of tea against chronic diseases (Klatsky et al., 1993; Hertog et al., 1997; Woodward and Tunstall-Pedoe, 1999). One of the reasons for this inconsistency may be that tea is not the only catechin-containing food. Including all sources of catechins in epidemiological studies may clarify the association between catechins, tea as one of its major sources, and chronic diseases. We have already reported on the catechin contents of a comprehensive set of fruits, vegetables, and certain staple and processed foods (Arts et al., 2000).

Although tea has been studied extensively for its biological actions, there are surprisingly few data on the catechin content of tea infusions. Previous studies typically determined the catechin contents of fresh tea leaves on a dry weight basis after exhaustive extraction with organic solvents. This type of data does not take into account infusion rates into ordinary hot water and is not suitable for use in epidemiological studies. The goal of the present study was to determine the catechin contents of commonly consumed beverages. We determined the following catechins, (+)-catechin, (−)-epicatechin, (+)-gallocatechin (GC), (−)-epigallocatechin (EGC), (−)-epicatechin gallate (ECg), and (−)-epigallocatechin gallate (EGCg) (Figure 1), in 8 types of black tea, 18 types of red and white wines, apple juice, grape juice, iced tea, beer, chocolate milk, and coffee.

EXPERIMENTAL PROCEDURES

Sample Collection and Preparation. *Tea.* The three most frequently consumed blends of black tea from two major brands and two fruit-flavored black teas were purchased (consumption data were obtained from the Coffee and Tea Information Bureau) and prepared according to Dutch custom. A 2-g tea bag was placed in 200 mL of boiled tap water for 5 min. Before the bag was removed, it was stirred through the brew a few times, after which the brew was allowed to cool. For the analysis of GC and EGC the pH of the brew was lowered by adding 50 μL of 1 M citric acid in methanol to 8 mL of tea, and the volume was made up to 10 mL with methanol (final pH 3). Lowering the pH stabilized these compounds in the HPLC autosampler for at least 24 h at room temperature. For the analysis of all other catechins the brew was diluted 10 times with methanol; stabilization was not necessary. The effect of the brewing method on catechin yield of Ceylon tea was studied by varying the infusion time (2, 5, and 10 min) and the amount of tea used for brewing (2, 2.5, 3, or 4 g).

Wine. Twelve red and six white wines commonly consumed in The Netherlands were purchased at an outlet of a nationwide supermarket chain that holds the major share of Dutch

* Address correspondence to this author at the State Institute for Quality Control of Agricultural Products (RIKILT), P.O. Box 230, 6700 AE Wageningen, The Netherlands (telephone +31 317 475578; fax +31 317 417717; e-mail p.c.h.hollman@rikilt.wag-ur.nl).

† State Institute for Quality Control of Agricultural Products (RIKILT).

‡ National Insitute of Public Health and the Environment (RIVM).

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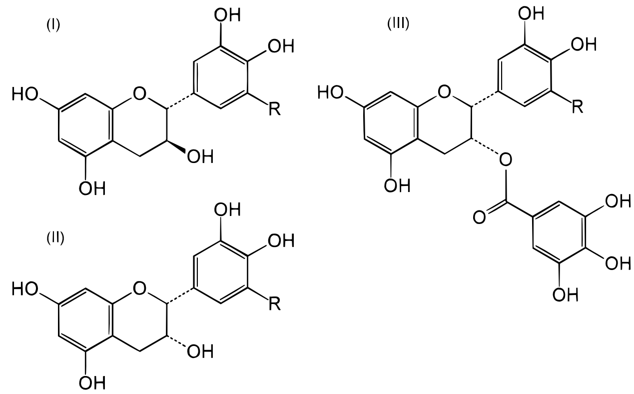

Figure 1. Chemical structures of catechins: (**I**) R = H (+)-catechin, R = OH (+)-gallocatechin (GC); (**II**) R = H (−)-epicatechin, R = OH (−)-epigallocatechin (EGC); (**III**) R = H (−)-epicatechin gallate (ECg), R = OH (−)-epigallocatechin gallate (EGCg).

wine sales (Albert Heijn). Types of wine were selected on the basis of information from the Dutch Commodity Board of Wine. Two types of French red wines (Bordeaux and Cotes-du-Rhone) were chosen. Of each, three brands (one low-priced, one medium-priced, and one higher-priced) were purchased. In addition, two Spanish, two South African, and two Italian red wines were selected. Three white Bordeaux wines and three German white Mosel-Saar-Ruwer wines in different price categories were purchased. Five milliliters of white wine was diluted with 5 mL of 90% methanol in water. Red wines were diluted 10 times by adding 18 mL of 90% methanol in water to 2 mL of wine.

Other Beverages. Two major brands (Riedel and Albert Heijn's own brand) of apple juice and black grape juice and one brand (Albert Heijn's own brand) of white grape juice were analyzed. Although orange juice is the most frequently consumed fruit juice in The Netherlands, it was not included because we did not detect catechins in oranges (Arts et al., 2000). One brand of iced tea (Liptonice lemon without carbon dioxide) was purchased, as was one brand of lager beer (Heineken). The beer was degassed in an ultrasonic bath at room temperature before injection. Two brands of commonly consumed semiskimmed chocolate milk (Albert Heijn's own brand and Nutricia), and a commonly consumed brand of coffee (Roodmerk, DE) were bought. The coffee was prepared according to Dutch custom in a coffee maker with a paper filter; 275 mL of boiling water was dripped on 14 g of finely ground coffee.

Analytical Methods. Six major catechins [(+)-catechin, (−)-epicatechin, GC, EGC, ECg, and EGCg] (Figure 1) were determined according to a method described previously in detail for solid foods (Arts and Hollman, 1998). Modifications were made to take the liquid sample matrix into account. After filtration over a 0.45 μm Acrodisc filter, the beverages were injected directly, without extraction or sample cleanup, onto an HPLC-UV-fluorescence system, operated at 30 °C. Prior to filtration, tea and wine were diluted as described. Chocolate milk required some cleanup because of its protein and fat content: 15 mL of chocolate milk was extracted for 1 h in a mechanical shaker with 5 mL of 1 M citric acid in methanol and subsequently centrifuged for 5 min at 2000*g*; the supernatant was injected. Quantification of (+)-catechin and (−)-epicatechin was done with fluorescence detection (280 nm excitation, 310 nm emission wavelengths), whereas the other compounds were measured with UV (270 nm). In addition to an acetonitrile/phosphate buffer gradient (Arts and Hollman, 1998), a second gradient using a methanol/phosphate buffer was applied (Arts et al., 2000).

Analytical Quality Control. A control sample of black tea infusion was included at the beginning and end of each series of analysis. For this control sample, the contents of 25 tea bags (2 g each) from different batches of one tea blend (Ceylon, Pickwick DE) were mixed, passed through a set of sieves retaining the >0.4 and <0.8 mm fraction, and stored in an airtight container. The control tea infusion was prepared fresh as follows: 200 mL of tap water was boiled, and 2 g of the control tea was added and allowed to infuse for 5 min. After infusion, the tea was passed through a household tea sieve, diluted similarly to the other tea samples, passed though a 0.45 μm Acrodisc filter, and injected. 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). All samples were analyzed in duplicate. Peak identification was done by comparing retention times and UV spectra obtained by diode array detection to those of pure standards. See the accompanying paper for details (Arts et al., 2000).

RESULTS AND DISCUSSION

Tea. Tea was the only beverage that contained EGCg (Table 1). Together with ECg it was in fact the most abundant compound present in black tea. (+)-Catechin and GC represented only a small part of the catechin content of our black teas, whereas (−)-epicatechin and EGC were low in English melange and Earl Grey but much higher in Ceylon tea. The total catechin content of Ceylon tea was very high (up to 418 mg/L) and approximated not only the composition of green tea (Khokhar et al., 1997) but also that of a mixture of orange Pekoe and Pekoe cut (Bronner and Beecher, 1998). The English melange and Earl Grey infusions analyzed in our study resembled black teas from India and the United Kingdom except for ECg and EGCg, which were higher in our tea brews (Khokhar et al., 1997). Because of the limited number of reports on catechin contents of tea infusions, we recalculated literature data on tea extracted with water, but with catechin contents expressed on a tea leaf dry weight basis, to enable comparisons to our data (Table 1). The large variation in contents of individual catechins in black tea brews from various origins is apparent. Tea blend, manufacturing practices, and methods of beverage preparation influence the composition of a tea brew (Graham, 1992). Regarding beverage preparation, it is apparent from our data that the quantity of tea used had more impact on the catechin concentration of the brew than the infusion time (Figure 2). In the range we tested (2−4 g/200 mL), catechin concentrations increased fairly linearly with the amount of tea used. On the other hand, maximum catechin concentrations were nearly reached after 5 min of infusion and did not increase substantially after that period. Clearly, if the composition of tea brews varies to the extent seen here, it may be difficult to interpret results from epidemiological studies on the health effects of tea consumption in an unambiguous way.

Wine. Wine contained only (+)-catechin and (−)-epicatechin (Table 2); levels in red wine [16.3−53.4 mg of (+)-catechin/L, 9.2−42.1 mg of (−)-epicatechin/L] were much higher than those in white wine [1.5−6.1 mg of (+)-catechin/L, 0.5−1.3 mg of (−)-epicatechin/L]. French red wines appeared to have higher levels of catechins than other red wines, which is consistent with reported data on the catechin content of 836 red wines from a large number of wine-making regions of the world (Goldberg et al., 1998). Goldberg and co-workers noted the large differences in catechin levels among wines made from different grape cultivars and the exceptionally high catechin content of wine made from Pinot Noir grapes (up to 287 mg/L). Pinot Noir wine was not included in our study because it is not frequently consumed in The Netherlands. Most of the wine from Burgundy, France, is made from this grape. Comparison of our data to those reported in the literature is difficult

Table 1. Catechin Content of Black Tea Infusions[a]

tea	catechin content (mg/L)[b]							reference
	(+)-catechin	(−)-epicatechin	GC	EGC	ECg	EGCg	total catechins	
English melange (Albert Heijn)	5.2	22.1	8.9	12.2	71.4	56.0	175.7	
English melange (Pickwick DE)	3.5	12.7	5.6	6.4	46.6	27.2	101.9	
Ceylon (Albert Heijn)	8.7	62.4	26.1	92.7	100.0	128.4	418.4	
Ceylon (Pickwick DE)	8.8	47.7	27.8	88.4	79.1	100.4	352.1	
Earl Grey (Albert Heijn)	6.0	13.5	6.1	2.9	70.5	51.3	150.3	
Earl Grey (Pickwick DE)	6.2	15.1	7.0	6.5	60.6	43.7	139.1	
lemon melange (Pickwick DE)	5.8	21.3	10.5	12.6	64.6	50.9	165.6	
forest fruit melange (Pickwick DE)	4.7	17.8	8.9	8.7	59.1	42.7	141.9	
literature data on black tea								
orange Pekoe/Pekoe cut (U.S.)	–[c]	40	–	60	110	120	330	Bronner and Beecher, 1998
Ceylon (The Netherlands)	–	79	–	91	85	229	484	Khokhar et al., 1997
Yule (India)	–	31	–	ND[d]	8	18	57	Khokhar et al., 1997
PG-tips (U.K.)	–	40	–	ND	10	26	76	Khokhar et al., 1997
black tea (Taiwan)[e]	–	9	–	116	7	32	164	Yen and Chen, 1996
black tea[e]	–	22	–	59	75	191	347	Kuhr and Engelhardt, 1991
black tea (China)[e]	120	54	–	52	349	40	615	Shao et al., 1995

[a] One tea bag (2 g) in 200 mL of boiling tap water during 5 min. [b] Average of duplicate analyses. [c] –, not determined. [d] ND, not detected. [e] Data have been recalculated from tea leaf dry weight to mg/L and corrected for brewing method to resemble 1 g of tea in 100 mL of water assuming linearity of infusion.

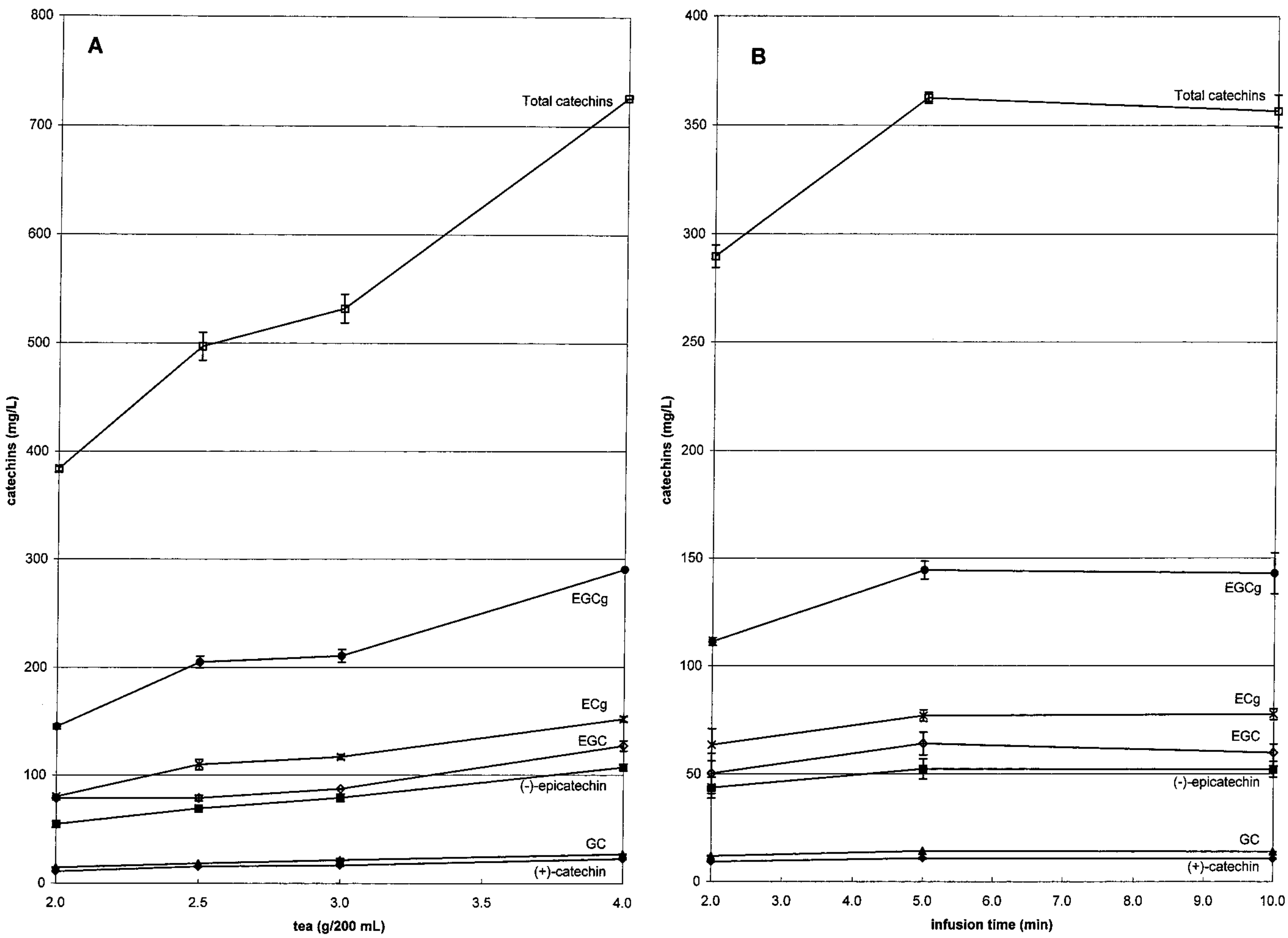

Figure 2. Catechin yield (milligrams per liter) with variations in brewing method: (A) amount of tea used for brewing; (B) infusion time. (□) Total catechins; (◆) (+)-catechin; (▲) GC; (■) (−)-epicatechin; (◇) EGC; (×) ECg; (●) EGCg. Values are means of duplicate analyses ± the standard deviation of the duplicates.

because grape cultivar has such a pronounced impact on the catechin concentration of wine. However, in general, results are in the same order of magnitude as ours (Etiévant et al., 1988; Archier et al., 1992; Soleas et al., 1997; Goldberg et al., 1998). Next to grape cultivar, which is the most important determinant of catechin content of wine, climatic conditions appear to play an important role. Within cultivars, the highest catechin levels were found in wines grown under damp cool conditions, whereas dry sunny climates yielded lower catechin concentrations (Goldberg et al., 1998). Even within France, an association was observed between the average temperature in a region and the catechin content of the wine produced there (Etiévant et al., 1988). These observations are consistent with our results: we found higher catechin levels in French wines

Table 2. Catechin Content of Red and White Wines, Fruit Juices, Beer, Chocolate Milk, and Coffee

product	catechin content (mg/L)[a]		
	(+)-catechin	(−)-epicatechin	total catechins
red wine			
Bordeaux (Appellation Controlee, Rineau, 1997)	53.4	42.1	95.5
Bordeaux Rouge (Appellation Controlee, Euroshopper, table wine)	47.7	41.5	89.2
Bordeaux (Appellation Controlee, Christian Moueix, 1993)	48.2	29.9	78.0
Cotes-du-Rhone (Appelation Controlee, Les Vendanges, 1997)	47.7	18.8	66.5
Cotes-du-Rhone (Appelation Controlee, Cuvee des Amoureuses, 1996)	44.5	19.0	63.6
Cotes-du-Rhone (Appelation Controlee, Reserve de l'Arche, 1996)	43.3	19.5	62.8
Rioja (Siglo, 1996)	22.5	9.2	31.7
Valdepenas (Albert Heijn's own brand, table wine)	29.0	13.2	42.1
Pinotage/Merlot (Stellenbosch Welmoed Winery, 1997)	25.2	18.9	44.1
Droe Rooi (Suid-Afrika, Albert Heijn's own brand, table wine)	16.3	11.0	27.3
Chianti Classico (Castellani, 1995)	23.2	16.2	39.4
Rosso del Salento (Albert Heijn's own brand, table wine)	21.6	16.4	38.1
white wine			
Bordeaux Sauvignon (Appellation Controlee, Chateau de Nivelle, 1997)	2.1	0.7	2.8
Bordeaux (Appellation Controlee, Rineau, 1997)	4.7	1.0	5.7
Bordeaux Sauvignon (Appellation Controlee, Euroshopper, table wine)	4.8	1.2	6.0
Mosel-Saar-Ruwer (Zeller Schwarze Katz, 1997)	3.6	1.3	4.8
Mosel-Saar-Ruwer (Qualitatswein, 1995)	1.5	0.5	2.0
Mosel-Saar-Ruwer (Riesling Spatlese Qualitatswein mit Pradikat, 1996)	6.1	0.8	6.8
other beverages			
apple juice (Riedel)	ND[b]	ND	ND
apple juice (Albert Heijn)	ND	ND	ND
black grape juice (Albert Heijn)	8.0	ND	8.0
black grape juice (Riedel)	9.1	ND	9.1
white grape juice (Albert Heijn)	2.0	ND	2.0
iced tea (Liptonice lemon without carbon dioxide)	ND	0.8	0.8
lager beer (Heineken)	ND	ND	ND
chocolate milk (semiskimmed, Nutricia)	16.1	5.0	21.1
chocolate milk (semiskimmed, Albert Heijn)	1.2	0.6	1.8
coffee (Roodmerk, DE)	ND	ND	ND

[a] Average of duplicate analyses; GC, EGC, ECg, EGCg were not detected in any of the samples. [b] ND, not detected.

than in Spanish and Italian wines. In our data, however, we cannot distinguish between cultivar and climate effects because we did not analyze wine from the same grape cultivar grown under different climatic conditions. Moreover, wine-making methods differ among regions as well and may explain some of the observed climatic differences.

Other Beverages. Despite the fact that both consumer apples (Risch and Herrmann, 1988; Arts et al., 2000) and homemade juice from apples used for commercial apple juice manufacture (Suárez Vallés et al., 1994) contain relatively high levels of (+)-catechin and (−)-epicatechin, no catechins were detected in commercial apple juices (Table 2). Previous studies likewise reported only very low levels of catechins in commercial apple juice [up to 2.3 mg of (+)-catechin and up to 0.7 mg of (−)-epicatechin/L of juice], or catechins were not detected (Bengoechea et al., 1997; Kermasha et al., 1995; Fernández de Simón et al., 1992; Spanos et al., 1990). Spanos and et al. (1990) showed that the complex process of commercial apple juice preparation results in a stepwise decrease in catechins. In particular, crushing and pressing, storage of the concentrated juice at room temperature, and decolorization by treatment with activated carbon are steps that lead to complete catechin loss. The same applies to grape juice: relatively high levels in homemade juice (Lee and Jaworski, 1987), whereas commercial juice, depending on the manufacturing method used, does not contain any catechins (Spanos and Wrolstad, 1990). In our study we found low levels of catechins in both white and black grape juices (Table 2).

In commercially available iced tea we detected very low levels of (−)-epicatechin (0.8 mg/L), whereas none of the other catechins normally present in tea were found. Iced ea is made from instant tea, and production methods of instant tea powder cause dimerization and polymerization of catechin monomers. Moreover, the amount of instant tea in commercially available iced tea is presumably low, and further degradation may have occurred during storage. The quantities of catechins in a number of instant tea powders ranged considerably, from 0.36 to 7.92% of dry weight (Constable et al., 1996). These data suggest that somewhat higher catechin levels may be found in other brands of iced tea. We did not investigate brands other than the most frequently consumed Dutch brand because iced tea consumption is not very high in The Netherlands.

Low levels of (+)-catechin (~5 mg/L) and (−)-epicatechin (~1 mg/L) have been reported in lager beers (Madigan et al., 1994; Achilli et al., 1993; Kaneda et al., 1990). We did not detect catechins in Dutch lager beer. Polyvinylpolypyrrolidone (PVPP) is an adsorbent for phenolics, which is used by breweries to prolong the stability of beers against haze formation. PVPP has been shown to reduce the catechin content of lager beer (McMurrough et al., 1995; Madigan et al., 1994). One might hypothesize that differences in the efficiency with which breweries clarify their beer explain the lack of catechins in the beer we analyzed.

Chocolate milk contained low levels of (+)-catechin and (−)-epicatechin. There was a large difference in catechin contents between the two brands tested. High levels of (+)-catechin and (−)-epicatechin have been reported previously in chocolate (Arts et al., 1999), cacao liquor (Sanbongi et al., 1998), and cacao beans (Porter et al., 1991; Kim and Keeney, 1984). The cacao content of chocolate milk is low, which results as a matter-of-course in low catechin levels. The difference in catechin content between the two brands tested may be due to different cacao contents of the beverage, but the use of cacao varieties with different initial catechin levels may be an alternative partial explanation.

Summary. Tea and red wine contained substantial quantities of catechins, whereas low to negligible amounts were found in white wine and commercially available fruit juices, iced tea, and chocolate milk. Catechins were absent from beer and coffee. Tea was the only beverage that contains all six catechins studied; only (+)-catechin and (−)-epicatechin were present in other beverages. Catechin contents may vary substantially among types, brands, and blends, particularly in tea. The data reported here, together with our data on solid foods (Arts et al., 2000), 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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