Content of the Flavonols Quercetin, Myricetin, and Kaempferol in 25 Edible Berries

Sari H. Häkkinen,[†,‡] Sirpa O. Kärenlampi,[§] I. Marina Heinonen,[∥] Hannu M. Mykkänen,[†] and A. Riitta Törrönen*[,†,‡]

Department of Clinical Nutrition, Department of Physiology, and Institute of Applied Biotechnology, University of Kuopio, P.O. Box 1627, FIN-70211 Kuopio, Finland, and Department of Applied Chemistry and Microbiology, P.O. Box 27, FIN-00014, University of Helsinki, Finland

The amounts of quercetin, myricetin, and kaempferol aglycons in 25 edible berries were analyzed by an optimized RP-HPLC method with UV detection and identified with diode array and electrospray ionization mass spectrometry detection. Sixteen species of cultivated berries and nine species of wild berries were collected in Finland in 1997. Quercetin was found in all berries, the contents being highest in bog whortleberry (158 mg/kg, fresh weight), lingonberry (74 and 146 mg/kg), cranberry (83 and 121 mg/kg), chokeberry (89 mg/kg), sweet rowan (85 mg/kg), rowanberry (63 mg/kg), sea buckthorn berry (62 mg/kg), and crowberry (53 and 56 mg/kg). Amounts between 14 and 142 mg/kg of myricetin were detected in cranberry, black currant, crowberry, bog whortleberry, blueberries, and bilberry. Kaempferol was detected only in gooseberries (16 and 19 mg/kg) and strawberries (5 and 8 mg/kg). Total contents of these flavonols (100−263 mg/kg) in cranberry, bog whortleberry, lingonberry, black currant, and crowberry were higher than those in the commonly consumed fruits or vegetables, except for onion, kale, and broccoli.

Keywords: *Flavonoid; flavonol; quercetin; myricetin; kaempferol; berry; fruit; HPLC*

INTRODUCTION

Flavonoids are polyphenolic phytochemicals that constitute a large group of secondary plant metabolites. These natural products are of interest because of their proposed health-promoting effects as antioxidants (Rice-Evans and Miller, 1998) and anticarcinogens (Strube et al., 1993). An inverse association between the intake of flavonols and flavones and the risk of coronary heart disease (Hertog et al., 1993a, 1995, 1997; Knekt et al., 1996), stroke (Keli et al., 1996), and lung cancer (Knekt et al., 1997) has been shown in epidemiological studies. In many countries, onions, apples, tea, and red wine are the main dietary sources of flavonoids (Hertog and Katan, 1998).

Berries are traditionally a part of the Finnish diet, because the northern location of Finland favors the growing of berries over fruits. We previously screened several berries for the flavonols kaempferol, quercetin, and myricetin and for selected phenolic acids (*p*-coumaric, caffeic, ferulic, *p*-hydroxybenzoic, and ellagic acids) (Törrönen et al., 1997; Häkkinen et al., 1998a) using a semiquantitative high-performance liquid chromatographic (HPLC) method (Häkkinen et al., 1998b). Although anthocyanins contribute a significant proportion of phenolic compounds in berries (Heinonen et al., 1998; Prior et al., 1998), they were not analyzed because the method was not suitable for that purpose. Flavonols represented >50% of the phenolics analyzed in sea buckthorn berries, crowberries, cranberries, lingonberries, and gooseberries (Törrönen et al., 1997; Häkkinen et al., 1998a). In black and red currants, bilberries, and chokeberries, the percentage of flavonols was >30%. On this basis it can be assumed that berries contribute a significant amount of flavonols to the Finnish or Nordic diet.

Flavonol contents of berries vary widely according to the literature. Herrmann and co-workers have studied flavonol contents in many berries using a combination of thin-layer chromatographic (TLC) and spectrophotometric methods (Wildanger and Herrmann, 1973; Starke and Herrmann, 1976). The amount of flavonols was 88 mg/kg (fresh weight) in black currants, 34 mg/kg in lingonberries, 29 mg/kg in raspberries, 2−27 mg/kg in red currants, and < 0.1 mg/kg in gooseberries. However, the extraction and hydrolysis procedures were not optimized for the compounds of interest. Similarly, flavonols in highbush blueberries (24−28.5 mg/kg, fresh weight) and cranberries (125−270 mg/kg) have been studied by Bilyk and Sapers (1986) by TLC methods with HPLC quantification but without optimization of extraction and hydrolysis (Bilyk et al., 1984). In the HPLC method of Hertog et al. (1992a,b), extraction and hydrolysis conditions for flavonols were optimized, but only three berries consumed in The Netherlands were studied. The contents of flavonols were 20.6, 3, and 249 mg/kg (fresh weight) in strawberries, red currants, and cranberries, respectively. Justesen et al. (1998) analyzed flavonols in seven berries consumed in Denmark using an HPLC method with photodiode array and mass spectrometric detection; among them were black currant (38 mg/kg, fresh weight), blueberry (73 mg/kg), cowberry (lingonberry) (215 mg/kg), and cranberry (380 mg/kg).

* Author to whom correspondence should be addressed (telephone +358 17 163109; fax +358 17 163112; e-mail riitta.torronen@uku.fi).
[†] Department of Clinical Nutrition.
[‡] Department of Physiology.
[§] Institute of Applied Biotechnology.
[∥] Department of Applied Chemistry and Microbiology.

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Flavonol glycosides were analyzed as rutin in several Californian berries by Heinonen et al. (1998). The apparent amounts of flavonols varied in blueberries (115−139 mg/kg, fresh weight), red raspberries (19−20 mg/kg), and strawberries (6.1−78 mg/kg) depending on the exctraction solution (60% methanol or 70% acetone) used.

Great variation in the flavonol contents may be explained by differencies in the berry varieties (Bilyk and Sapers, 1986; Amiot et al., 1995) or growth conditions (Dixon and Paiva, 1995). Also, methodological differencies (Hertog et al., 1992a; Häkkinen et al., 1998b; Heinonen et al., 1998) may partly explain the great variability in the apparent flavonol contents reported. This variation, however, makes the evaluation of the berries as dietary sources of flavonols difficult. In addition, knowledge of the flavonol content of some berries consumed in Finland (e.g., cloudberry, sea buckthorn berry) is lacking.

The aim of the current study was to investigate the amounts of the flavonols quercetin, myricetin, and kaempferol in edible berries growing and commonly consumed in Finland. An optimized HPLC method with UV detection was used for quantification, and the flavonol aglycons were identified by their UV spectra (diode array detector, DAD) and mass spectra (electrospray ionization mass spectrometry, ESI-MS) (Häkkinen and Auriola, 1998). As berries are known to be good sources of another type of antioxidant, vitamin C (Rastas et al., 1997), the contents of this vitamin in the berries were determined, and the correlation between flavonol and vitamin C contents was calculated. Finally, an estimation of the contribution of berries to the dietary intake of flavonols in Finland was calculated using food consumption data from the Finnish Household Survey 1990 (Statistics Finland, 1993).

EXPERIMENTAL PROCEDURES

Berry Samples. Sixteen species of cultivated berries and nine species of wild berries were studied. They were of Finnish origin and collected in 1997 from eastern Finland unless otherwise stated. Wild crowberries were also collected from several places in Lapland and pooled before analysis. Samples of wild cloudberries, bilberries, lingonberries (cowberries), and cranberries were also collected from western Finland. Fresh berries were frozen and stored for 3 months at −20 °C until analyzed.

Thawing of the Frozen Berries. The influence of thawing method was tested for three berries (strawberry, bilberry, and lingonberry). All analyses were carried out in triplicate. The berries were thawed in a refrigerator (7 °C, 16 h), at room temperature (21 °C, 1.5 h), or in a microwave oven (2−3 min) in plastic containers. In all cases the berries were cold (5−10 °C) when homogenized.

Extraction and Hydrolysis. Flavonols were extracted and hydrolyzed to aglycons with a method modified from that of Hertog et al. (1992a). The procedure used is described in detail in Häkkinen and Auriola (1998). The berry samples (5 g) were refluxed for 2 h at 85 °C in 50% (v/v) aqueous methanol containing hydrochloric acid (1.2 M) and *tert*-butylhydroquinone (TBHQ) as an antioxidant.

In a preliminary experiment, extraction solutions with methanol concentrations of 25, 50, and 64% were tested in the analysis of flavonols from lingonberry and bilberry. In all extractions, 1.2 M HCl was used. Also, the influence of TBHQ on flavonol concentrations was investigated.

Chemicals. Quercetin and kaempferol were purchased from Sigma Chemical Co. (St. Louis, MO). Myricetin was obtained from Fluka Chemie AG (Buchs, Switzerland). The standards were dissolved in methanol. TBHQ was obtained from Aldrich-Chemie (Steinheim, Germany). Methanol (Lab-Scan, Dublin, Ireland) and acetonitrile (Rathburn, Walkerburn, Scotland) were of HPLC grade. Formic acid (Merck, Darmstadt, Germany) and hydrochloric acid (Riedel-deHaën, Seelze, Germany) were of analytical grade.

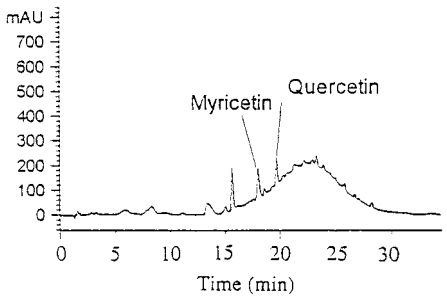

Figure 1. Typical chromatogram of a black currant extract monitored in 1% formic acid/acetonitrile gradient. Detection at 360 nm; flow rate = 0.5 mL/min.

Apparatus. The HPLC system used for UV quantification was a Hewlett-Packard (Waldbronn Analytical Division, Germany) instrument with a 1050 Series quaternary pump, an autosampler, and a variable-wavelength detector. For the identification of flavonols in berries, an HPLC apparatus consisting of a Hewlett-Packard 1050 Series pumping system, an injector, and a 1040M Series II diode array UV−vis detector was used. The system used for HPLC/MS analyses was a Finnigan MAT LCQ ion trap mass spectrometer (San Jose, CA) equipped with a Rheos 400 HPLC pump (Danderyd, Sweden).

Chromatographic Systems. The columns used and the identification procedures are described in detail in Häkkinen and Auriola (1998). The gradient was first optimized using a mixture of flavonol (kaempferol, quercetin, and myricetin) and phenolic acid (*p*-coumaric, caffeic, ferulic, and ellagic acid) standards. The reason for testing both phenolic acids and flavonols together was that, in our earlier studies, ellagic acid and myricetin were not adequately separated from each other in some elution systems (Häkkinen et al., 1998b). After testing with three hydrolyzed berry extracts, the final gradient was as follows: 0−10 min, 10−13% of B in A; 10−25 min, 13−70% of B in A; 25−29 min, 70% of B in A; 29−30 min, 70−10% of B in A; 30−35 min, 10% of B in A. Solvent A was 1% formic acid and B was acetonitrile. By using this gradient (flow rate = 0.5 mL/min), best purity and separation were achieved for flavonol peaks in black currant (Figure 1), lingonberry, and strawberry.

Analytical Quality Control. To study the within-laboratory repeatability (within-day precision), the flavonol content of a frozen black currant sample was analyzed six times within 1 day. Within-laboratory reproducibility (day-to-day precision) was studied by duplicate analyses of a freeze-dried bilberry sample on 9 days during a period of 7 months. The control sample was stored at −20 °C between the analyses.

Every week, new standard curves with freshly prepared standards (2, 60, and 300 ng in 1 μL of methanol) from stock solutions were determined. The stock solutions of the standards in methanol (1000 μg/mL) were stable for at least 2 months at −20 °C.

Recoveries were measured in each berry species (except for green currant, white currant, blueberries, and sweet rowan, for which the recoveries of black currant, red currant, bilberry, and rowanberry were used, respectively) by spiking the extraction solutions with pure flavonols at the level of 50−100% of the measured content, prior to sample analysis. Recoveries are shown in Table 1, and data reported in this paper have been corrected accordingly.

Analysis of Vitamin C. After partial thawing at room tempereture, the frosty berries (100 g) were quickly homogenized in a food processor, and a 20-g sample of the homogenate was weighed. Ice-cold 1.5% (w/v) metaphosphoric acid, pH 3.5−4.0 (200 mL), was immediately added. Triplicate samples of each berry were prepared. A colorimetric, enzymatic

Table 1. Recoveries (Percent) of Pure Flavonols Added to the Berry Samples ($n = 3$) Prior to Extraction and Hydrolysis

berry	quercetin	myricetin	kaempferol
family Grossulariaceae			
black currant[a]	50	52	
red currant	93		
gooseberry	54		67
family Ericaceae			
bog whortleberry	84	83	
lingonberry[a]	50		
cranberry	65	58	
bilberry[a]	86	70	
family Rosaceae			
strawberry	88		88
chokeberry	106		
rowanberry	63		
red raspberry	94		
cloudberry	77		
arctic bramble	89		
family Empetraceae			
crowberry	93	99	
family Elaeagnaceae			
sea buckthorn berry	66		

[a] Recovery data analyzed from six samples.

method commercially available for the determination of L-ascorbic acid in foodstuffs and other materials (catalog no. 409 677, Boehringer Mannheim GmbH, Mannheim, Germany) was used. L-Ascorbate and other reducing substances reduce the tetrazolium salt MTT in the presence of an electron carrier to a formazan, which is measured photometrically at 578 nm. L-Ascorbate is oxidatively removed by ascorbate oxidase, and the absorbance difference in the absence and presence of the enzyme is equivalent to the quantity of L-ascorbate in the sample. Pure L-ascorbic acid (Riedel-deHaën) in metaphosphoric acid was used for quality control.

Calculation of Intake. The dietary intake of flavonols was calculated by using food consumption data obtained from the Finnish Household Survey 1990 (Statistics Finland, 1993). The survey recorded the purchase of foods (250−300 food items) for the household. The population of the survey comprised all private households in the country, from which a random sample of 12000 households was drawn. In 1990 the final sample size was 8258 households. Data were collected by interviews and by using food purchasing diaries kept for 2 weeks. Meals eaten outside the household were not recorded in detail.

Fresh berries recorded in the Finnish Household Survey were black currant, red and white currants, strawberry, other garden berries, bilberry, lingonberry, cranberry, and other wild berries. Consumption figures given in liters were converted to kilograms. For other flavonol-rich foods, the flavonol contents reported by Hertog et al. (1992b, 1993b) were used.

RESULTS AND DISCUSSION

Selection of the Thawing Method. Variability in apparent quercetin contents differed depending on the method of thawing of the frozen berries, being 3, 12, and 17% for thawing at room temperature, in the microwave oven, and in the refrigerator, respectively. Thawing at room temperature resulted in lower apparent quercetin contents in black currant and strawberry (24 and 5 mg/kg, respectively) than thawing in the microwave oven (29 and 6 mg/kg, respectively). Myricetin contents and variations were similar after thawing at room temperature or in the microwave oven. Kaempferol content of strawberries was independent of the thawing conditions. On the basis of these results, thawing in the microwave oven (2−3 min) was chosen. This was also the most practical method for routine analysis.

Optimization of Extraction and Hydrolysis. When extracted with 50% aqueous methanol (the final method), the apparent flavonol contents in the berries were 20 or 70% higher than those recovered with 64 or 25% methanol, respectively. Also, Hertog et al. (1992a) reported that the apparent flavonoid levels in onion, endive, and celery were up to 30% higher when 50% instead of 80 or 20% aqueous methanol was used.

In lingonberry, the apparent quercetin content increased by 8% when TBHQ (50 mg/50 mL) was present in the extraction and hydrolysis solution. In bilberry, myricetin and quercetin contents were 20 and 30% higher in the presence of TBHQ, respectively. Therefore, TBHQ was used as an antioxidant in the flavonol analyses.

Validation of the Method. Studied on selected berries, the coefficients of variation (CV) for the repeatability (from frozen black currants) and within-laboratory reproducibility (from freeze-dried bilberries) of the HPLC method were 9.3 and 9.2% for quercetin and 6.1 and 9.4% for myricetin, respectively. Hertog et al. (1992a) reported a slightly better repeatability (CV < 5%) and reproducibility (CV < 7%) from freeze-dried cranberry.

Injection of berry extracts into HPLC has to be done immediately after the evaporated sample is dissolved in methanol. The apparent quercetin content of lingonberry decreased by 30% during 3 h in methanol, and in bilberry and black currant by 30 and 50%, respectively, during 7 h. Myricetin concentration decreased by 10 and 30% in bilberry and black currant, respectively, during 7 h of storage in methanol. In the standard solutions, quercetin, myricetin, and kaempferol concentrations were stable for 8 h; CVs for 16 injections were 4, 3, and 3%, respectively.

Recovery of pure quercetin ranged from 50 to 106%, that of myricetin from 52 to 99%, and that of kaempferol from 67 to 88% (Table 1). Because of the differences between berries, the results were corrected for the recovery. Good recoveries (95−105%) were found when pure flavonol aglycons were spiked into the extraction and hydrolysis solution without berries. The influence of some berry matrices was detrimental for the analysis of some flavonol aglycons. This may be due to their chemical reactions as metal chelators or to copigmentation reactions with other phenolics, for example, anthocyanidins (Britton, 1983). Most likely, enzymatic reactions did not cause the poor recoveries of flavonols in some berries, because methanol (50% v/v) and HCl (1.2 M) in the extraction medium denaturate plant enzymes such as polyphenol oxidase (Markham and Bloor, 1998).

Flavonol Contents. Table 2 shows the flavonol contents of Finnish berries harvested in 1997. Two values are reported whenever the berries were collected from two different geographical locations. Great differences among the berries were found in their flavonol contents (from 6 to 263 mg/kg, fresh weight). Similar levels of total flavonols were found among cultivars and strains in all cases when such comparisons could be made (strawberry, red raspberry, blueberry, and crowberry). The major flavonol found was quercetin; in only two berries (black currant and cranberry) was the content of myricetin higher than that of quercetin. Myricetin was also detected in bog whortleberry, crowberry, blueberries, and bilberry. Kaempferol could only be detected in gooseberries and strawberries.

Table 2. Flavonol and Vitamin C Contents of Finnish Berries (Milligrams per Kilogram, Fresh Weight)

berry	scientific name	quercetin[a]	myricetin[a]	kaempferol[a]	total flavonols	vitamin C[a]
family Grossulariaceae						
black currant	*Ribes nigrum* Öjebyn	44	71	nd	115	904
green currant	*Ribes nigrum* Vertti	32	nd	nd	32	846
red currant	*Ribes* × *pallidum* Red Dutch	9	nd	nd	9	187
white currant	*Ribes x pallidum* White Dutch	7	nd	nd	7	189
gooseberry, yellow	*Ribes uva-crispa*	18	nd	16	34	174
gooseberry, red	*Ribes uva-crispa*	22	nd	19	41	256
family Ericaceae						
bog whortleberry	*Vaccinium uliginosum* (wild)	158	26	nd	184	532
lingonberry[b]	*Vaccinium vitis-idaea* (wild)	74, 146	nd	nd	74, 146	75
cranberry[b]	*Vaccinium oxycoccos* (wild)	83, 121	74, 142	nd	157, 263	200
bilberry[b]	*Vaccinium myrtillus* (wild)	29, 30	14, 21	nd	43, 51	nd
blueberry	*Vaccinium corymbosum* Northcountry	24	26	nd	50	nd
	Vaccinium corymbosum Northblue	17	23	nd	40	nd
family Rosaceae						
strawberry	*Fragaria* × *ananassa* Senga Sengana	7	nd	8	15	478
	Fragaria × *ananassa* Jonsok	7	nd	5	12	420
chokeberry	*Aronia mitschurinii* Viking	89	nd	nd	89	na
rowanberry	*Sorbus aucuparia* (wild)	63	nd	nd	63	490
sweet rowan	*Grataegosorbus mitschurinii* Granatnaja	85	nd	nd	85	121
red raspberry	*Rubus idaeus* Ottawa	8	nd	nd	8	252
	Rubus idaeus Muskoka	8	nd	nd	8	272
	Rubus idaeus (wild)	6	nd	nd	6	na
cloudberry[b]	*Rubus chamaemorus* (wild)	6, 6	nd	nd	6, 6	697
arctic bramble	*Rubus arcticus* Pima and Mespi	31	nd	nd	31	208
family Empetraceae						
crowberry[c]	*Empetrum hermaphroditum* (wild)	53	49	nd	102	37
	Empetrum nigrum (wild)	56	44	nd	100	na
family Elaeagnaceae						
sea buckthorn berry	*Hippóphaë rhamnoides*	62	nd	nd	62	803

[a] Mean of triplicate assays from edible part; flavonol data have been corrected for recoveries (Table 1). nd, not detectable; na, not analyzed. [b] The two values represent analyses from berries collected from two different parts of Finland. [c] *E. hermaphroditum* sample was a pool of berries collected from three different places in Lapland.

In general, the highest quercetin concentrations were found in wild berries of the families Ericaceae (17−146 mg/kg), Empetraceae (53 and 56 mg/kg), and Eleagnaceae (62 mg/kg). Members of the family Grossulariaceae had moderately high quercetin contents (18−44 mg/kg), except for red and white currants. The berries of the genera *Rubus* and *Fragaria* of the family Rosaceae had low quercetin contents (6−8 mg/kg); however, the other members of the family Rosaceae (chokeberry and rowanberries) had high quercetin contents (31−89 mg/kg). These data are in accordance with our previous data (Törrönen et al., 1997; Häkkinen et al., 1998a) showing certain similarities within families and genera in the phenolic profiles of berries.

Compared to our results, Herrmann and co-workers (Wildanger and Herrmann, 1973; Starke and Herrmann, 1976) measured lower flavonol levels for lingonberries, gooseberries, and black currants. Wildanger and Herrmann (1973) reported similar results for quercetin in bilberries. In addition, we also detected myricetin in blueberries and bilberries (14−26 mg/kg). Differences in flavonol contents may partly be due to different cultivars or varieties in Finland and in Germany or to methodological differences. Also, environmental factors (e.g., light, temperature, and soil nutrients) may influence phenylpropanoid metabolism and flavonol concentrations in plants (Dixon and Paiva, 1995). Discrepancies may also be due to differences in maturity stage of the analyzed fruit (Amiot et al., 1995; Prior et al., 1998). Our results for quercetin contents in red and white currants are within the range found in red and white currant cultivars by Herrmann and co-workers (Wildanger and Herrmann, 1973; Starke and Herrmann, 1976).

The results of Bilyk and Sapers (1986) on American highbush blueberry cultivars are in good agreement with our data on quercetin. However, in contrast to their results, we also found myricetin (23 and 26 mg/kg) in blueberries. In wild cranberry, higher levels of myricetin and similar levels of quercetin were found in the present study compared to those of many different cranberry varieties by Bilyk and Sapers (1986). These discrepancies may be due to differences in varieties or cultivars; according to Bilyk and Sapers (1986), the variation in quercetin (112−250 mg/kg) and myricetin (11−24 mg/kg) contents among six cranberry varieties was large.

Our results for strawberries, red currants, and red raspberries are in good accordance with flavonol levels reported by Hertog et al. (1992b) and Justesen et al. (1998). In cranberries, higher levels of quercetin and lower levels of myricetin were reported by Hertog et al. (1992a). Different results might be explained by different varieties, for example, *Vaccinium macrocarpon* Ait. versus *Vaccinium oxycoccos*. Flavonol contents in lingonberries (cowberries), cranberries, and blueberries were lower in the present study than those reported by Justesen et al. (1998). In black currant, we found a content of quercetin similar to that reported by Justesen et al. (1998). In addition, we also detected myricetin in black currant (71 mg/kg). Heinonen et al. (1998) found in California blueberries, red raspberries, and strawberries higher flavonol concentrations than were found in the present study. This might be due to varietal differences or to the use of acetone in extraction procedure; acetone was superior to methanol in extracting phenolic compounds from the berry material without hydrolysis (Heinonen et al., 1998). To our knowledge, no data on flavonol content in cloudberry, arctic bramble, sea buckthorn berry, bog whortleberry, or crowberry have been previously reported, except for our semiquantitative studies (Törrönen et al., 1997; Häkkinen et al., 1998a).

Table 3. Intake of Flavonols in Finland in 1990

foodstuff	annual consumption[a] (kg or L[b])	flavonol content (mg/kg or mg/L[b])	intake (mg/day)
unprocessed berries			
black currants	1.25	115	0.39
red and white currants	0.88	9	0.02
strawberries	4.00	15	0.16
bilberries	1.39	47	0.18
lingonberries and cranberries[c]	1.33	130	0.47
cloudberries	1.24	6	0.02
other foodstuffs and beverages			
onions	3.08	347[d]	2.97
tomatoes	7.13	8[d]	0.16
apples	10.78	36[d]	1.06
grapes	1.21	13[d]	0.04
juice drinks (as orange juice)	20.95	5[d]	0.26
tea[e]	16	35[d]	1.53
red wine[f]	0.5	16[d]	0.02
total			7.28

[a] Statistics Finland (1993). [b] Juices, tea, wine. [c] Consumption ratios of lingonberries and cranberries are estimated to be 80 and 20%, respectively. [d] Contents of flavonols from Hertog et al. (1992b, 1993b). [e] Tea drink prepared from 0.16 kg of tea leaves. [f] Twenty percent of the consumption of mild wines (2.47 L).

The location where the berries were collected had no influence on quercetin levels in bilberry and cloudberry. Myricetin content in bilberries from western Finland was, however, lower than that in bilberries from eastern Finland. In lingonberries collected from two different parts of Finland, quercetin contents differed markedly (74 and 146 mg/kg). Cranberries collected from western Finland had less flavonols (157 mg/kg) than those collected from eastern Finland (263 mg/kg). The location effect may partly explain this difference. Cranberries with the lower flavonol content had also been exposed to night frost, and the maturities of the berries may have been different, although both were ripe. In the study of Starke and Herrmann (1976), quercetin and myricetin glycosides in black currants increased considerably during ripening. However, in other berries (e.g., red and white currants, cultivated blueberries, and sour cherries) the levels of kaempferol and quercetin glycosides were lower in ripe than in unripe fruits.

Cranberry, bog whortleberry, lingonberry, black currant, and crowberry had higher total flavonol contents (100−263 mg/kg) (Table 2) than do other fruits or most vegetables (Hertog et al., 1992b). Only onions, kale, and broccoli contain similar or higher amounts of flavonols (102−347 mg/kg). Chokeberry, rowanberry, sea buckthorn berry, bilberry, blueberry, gooseberry, green currant, and arctic bramble had similar or higher flavonol levels than apples (36 mg/kg) have (Hertog et al., 1992b).

Vitamin C Contents. Vitamin C concentrations (Table 2) did not correlate with total flavonol concentrations in the berries studied ($r = 0.02$, $n = 22$). In general, vitamin C concentrations were higher than flavonol concentrations, with the exception of lingonberry, cranberry, crowberry, bilberry, and blueberries. In black currant, sea buckthorn berry, and bog whortleberry the contents of both total flavonols and vitamin C were high (>50 and >500 mg/kg, respectively).

Flavonol Intake. The total annual consumption of unprocessed berries in 1990 in Finland was 10 kg/person, providing 1.24 mg flavonols/day (Table 3). Lingonberries and cranberries (annual consumption = 1.33 kg) provided 38% of the total flavonol intake from berries. Although strawberries represented 40% of the total berry consumption, their contribution to the flavonol intake from berries was only 13%. It should be recognized that the intake of flavonols from berries harvested in different growing seasons may vary. Because the flavonol data were obtained from berries collected during one growing season only, our estimation of intake should be considered tentative.

The intake of flavonols from meals eaten at home was 7.28 mg/day (Table 3). The main sources were onions (41%), tea (21%), berries (17%), and apples (15%). Assuming that one-third of the meals was eaten outside the home, the intake of flavonols would have been ~11 mg/day. Thus, the flavonol intake in Finland was in 1990 almost twice the intake (6 mg/day) 40 years ago reported by Hertog et al. (1995) and 3 times the flavonoid intake (3−4 mg/day) ~30 years ago reported by Knekt et al. (1996, 1997). In these studies (Hertog et al., 1995; Knekt et al., 1996, 1997), methods different from that in the present study were used for the collection of food consumption data.

In this study the emphasis was on the analysis of flavonols. In addition to flavonols, berries also contain significant quantities of other phenolic compounds such as anthocyanins (Heinonen et al., 1998; Prior et al., 1998) and phenolic acids (Törrönen et al., 1997; Heinonen et al., 1998; Häkkinen et al., 1998a,b). Intake of these phenolics from berries is presumably much higher than that of flavonols.

Conclusions. In this study, three flavonols were identified and quantified from various berries. According to Hertog et al. (1995), the flavonol content of food is high if it is >50 mg/kg. On this basis, the flavonol content was high in 12 berries (cranberry, bog whortleberry, lingonberry, black currant, two species of crowberries, chokeberry, sweet rowan, rowanberry, sea buckthorn berry, bilberry, and blueberry var. Northcountry). Of the berries studied, only cloudberries, red raspberries, and red and white currants had low levels of flavonols (<10 mg/kg). The unprocessed berries make a moderate contribution (17%) to the total dietary intake of flavonols in Finland. A reasonable portion (100 g) of lingonberries or black currants would increase daily flavonol intake by 100% and that of bilberries by 45%. Thus, the berries commonly consumed in Finland are excellent potential sources of dietary flavonols. Precise knowledge of the flavonol contents of berries is important to assess the effect of this class of compounds on human health and disease.

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