

Content of Selected Flavonoids in 100 Edible Vegetables and Fruits

Jie CAO, Wei CHEN, Ying ZHANG, Yuqiu ZHANG and Xiujuan ZHAO *

Department of Nutrition and Food Hygiene, Public Health School, Harbin Medical University, 194 Xuefu Road, Harbin Heilongjiang P.R.China

Received August 4, 2009; Accepted May 10, 2010

The objective of this work was to quantify the potential anticancer flavonoids (quercetin, kaempferol, isorhamnetin, apigenin and luteolin) by a rapid, reliable and accurate reverse phase high performance liquid chromatography (RP-HPLC) among 100 edible vegetables and fruits commonly consumed in Harbin city, China. The predominant flavonoids found in the largest abundance in all of the analyzed foods were kaempferol, followed by luteolin and quercetin. The maximum content of quercetin, kaempferol, isorhamnetin, apigenin, luteolin and total flavonoids in foods amounted to $379.7 \pm 19.7 \text{ mg kg}^{-1}$, $482.0 \pm 21.0 \text{ mg kg}^{-1}$, $333.6 \pm 14.4 \text{ mg kg}^{-1}$, $139.3 \pm 5.2 \text{ mg kg}^{-1}$, $179.5 \pm 10.6 \text{ mg kg}^{-1}$ and 853.0 mg kg^{-1} fresh weight (FW), respectively. Potherb mustard, lettuce (red), toona leaf, celery leaf, garlic, garlic stalk, apple, date, ginger (with peel) and radish leaf (red root, small) were major food sources of flavonols and flavones. This study contribute to the establishment of China's flavonoid database and provide a basis for the assessment of the daily flavonoid intake.

Keywords: HPLC, vegetables, fruits, flavonols, flavones

Introduction

Flavonoids, which represent one of the most attracting phytochemical subgroups, are widespread in plant and plant-derived foodstuff such as vegetables, fruits, tea and wine (Scalbert *et al.*, 2000). Flavonoids can be subdivided into six categories according to their chemical structure. The subclasses and their respective food sources are flavones (green leafy species), flavonols (ubiquitous in foods), flavanones (citrus), anthocyanidins (red, purple and blue berries), flavanols (teas, red grapes and red wines) and isoflavones (soybeans) (i). Except for catechins, most of the compounds frequently occurring forms are the glycoside derivatives in plants. Within the subgroups of flavonols and flavones, the flavonol quercetin is the most frequently occurring compound in foods. Also common are kaempferol, isorhamnetin, and the flavones apigenin and luteolin.

Flavonoids have a wide range of biochemical and pharmacological effects, including antiinflammatory, antioxidant and antiproliferative actions, which may protect the body from various diseases, such as cancers and cardiovascular disease (Middleton *et al.*, 2000; Nijveldt *et al.*, 2001). For

example, flavones and flavonols may protect against renal cell carcinoma (Bosetti *et al.*, 2007), and increased the intake of flavones and flavonols may lower the risk of colorectal cancer (Rossi, *et al.*, 2006). To better understand the association between the intake of specific flavonoids and disease risk at the population level, precise analysis of the flavonoid intake are required.

Recently, the intake of flavonoids has been assessed in a variety of countries, from 20 mg/d in the United States to >70 mg/d in Holland (Beecher, 2003), from an analysis of the population's commonly consumed foods, such as vegetables, fruits or beverages. However, there are no data about the estimate of the daily flavonoid intake in China. The reason may be that there is not any available food composition database which could provide quantitative information on specific compounds in specific food. So far, little attention has been paid in China to quantify the content of flavonoids in foods. A large number of studies reported for the quantitative determination of flavonoids were either to determine a single flavonoids in a variety of foods or to determine all of the flavonoids in a single food. Although data such as the USDA database for flavonoids enables and facilitates the assessment of the daily flavonoid intake (Scalbert *et al.* 2000; Kinoshita *et al.*, 2005; Peterson *et al.*, 2000), the content of

*To whom correspondence should be addressed.

E-mail: xiujuan_zhao@sina.com

flavonoids in foods are strongly influenced by variations in cultivar, growing location, agricultural practices and processing and so on, and different populations in different countries consume different kinds of plant foods.

Therefore, in the present study, we quantified the content of the following flavonoids in plant species, three major flavonols quercetin, kaempferol and isorhamnetin and two major flavones luteolin and apigenin, which are most widely investigated in anticarcinogenesis studies. This work reported both the composition and content of flavonoids in 100 edible vegetables and fruits commonly consumed in Harbin city, China. In addition, we investigated the effect of season factors on flavonoid amounts in foods.

Materials and Methods

All chemicals were analytical grade or chromatography grade. The standards of kaempferol, quercetin, isorhamnetin, apigenin and luteolin were obtained from Sigma (St Louis, MO, USA).

The foods (vegetables and fruits) to be analyzed were selected for sampling on the basis of their high consumption (The consumption data of vegetables and fruits presented in Table 1 were obtained from our dietary survey for 5166 residents of Harbin.), a lack of data, and their expected flavonoid content. Vegetables and fruits sampled were all grown in China. Fresh samples were collected from retail outlets in eight regions of Harbin city. We chose three regions as

sampling sites from eight regions following the method of random number table. Afterwards, we chose at random three retail outlets as sampling sites from all retail outlets in each region with the method of random number table. Each regional sample was a composite of samples from three retail outlets. All vegetables and fruits (1–2 kg each) were generally purchased only as fresh products and at the time of their most frequent consumption during four periods (spring, 2008; summer, 2008; autumn, 2008; and winter, 2008). This approach was designed to ensure that analytical results were representative of the food supply.

After purchased, the nonedible parts of the samples were removed, and the edible parts were immediately cleaned, chopped into small pieces and mixed, and later freeze-dried, and stored at -20°C prior to analysis. At the time of analysis, all of the samples were finely powdered and passed through a 40 mesh sieve prior to extraction.

Hydrolysis and extraction were performed according to the method (Cao *et al.*, 2008). In brief, we got the freeze-dried samples 2.0 g, added with 80% ethanol/water/HCl 100 mL (80:10:10, v/v/v) and 0.20 g butylated hydroxy toluene (BHT), and then refluxed at 90°C for 3 h, cooled down to room temperature, filtered with Bucher, centrifuged at 3000 rpm for 20 min, approximately 2 mL supernatant was taken and filtered through 0.22 µm syringe filter (Millipore) for subsequent HPLC analysis.

Identification and quantification of flavonoid aglycons

Table 1. The estimated daily intake (g) of vegetables and fruits obtained in 5166 volunteers of Harbin city in China.

Food Description	Intake	Food Description	Intake
Red fuji apple	59.65	Coriander	15.12
Orange	46.47	Shaddock	13.54
Tomato	41.42	Kiwi fruit	13.24
Banana	37.89	Hot pepper, green	12.77
Cucumber	36.48	Mung bean sprouts	12.24
Watermelon	36.01	Garlic flowering stalk	11.74
Chinese cabbage	28.91	Carrot	11.08
Eggplant	25.81	Rape	10.90
Pear	24.55	Chive	10.22
Muskmelon	23.08	Pineapple	10.15
Orange, Navel	21.60	Romaine lettuce	9.57
Peach	19.46	Celery	9.28
Kidney bean, green	18.74	Date	8.69
Green pepper	18.11	Cauliflower	8.69
Spinach	16.85	Ginger	8.46
Onion	16.58	Zucchini, green	7.95
Sweet potato	16.40	Radish	7.49
Cabbage	16.40	Soybean sprouts	6.35
Grape	16.25	Pumpkin	4.96
Garlic	15.21	Chinese wax gourd	4.75

were achieved using the reversed-phase HPLC system (Cao, *et al.*, 2008) equipped with a Waters 2690 separation module and an ODS column (SYNERGI POLRR - RP80A, 250 mm \times 4.60 mm) at 30°C using isocratic elution. The column elute was monitored using a Waters 996 photo diode array detector (quercetin: 366.9 nm, kaempferol: 363.5 nm, isorhamnetin: 365.7 nm, apigenin: 338.6 nm, luteolin: 350.5 nm). Elution solvents (40/60, v/v) consisted of A (0.1% phosphoric acid aqueous solution) and B (methanol). A flow rate of 1ml/min was applied and 10 μ L of the final solution was injected.

Samples were injected in three times, and flavonoids were quantified using the respective external standards. Peak identification was performed by comparison of retention times and diode array spectral characteristics with the standards and the library spectra. Analytical performances of the method were compared in terms of linearity, limits of detection (LODs) and precision. Celery leaf containing various flavonoid aglycones analyzed was used as a control sample to determine individual flavonoid recovery. Mean recovery of flavonoid standards added to extraction solution ($n=3$) amounted to 97.90% (quercetin), 97.77% (kaempferol), 93.57% (isorhamnetin), 93.40% (luteolin), and 94.97% (apigenin). The method showed a good reproducibility with coefficients of variation of 2.50% (quercetin), 3.74% (kaempferol), 4.24% (isorhamnetin), 2.61% (luteolin), and 2.25% (apigenin). The detection limits of kaempferol, quercetin, isorhamnetin, apigenin and luteolin were 2.5 mg kg⁻¹ FW, 1.5

mg kg⁻¹ FW, 3.0 mg kg⁻¹ FW, 2.0 mg kg⁻¹ FW and 1.5 mg kg⁻¹ FW, respectively.

Results and Discussion

Analytical results Following the hydrolysis procedure, only aglycones were detected, so all concentrations reported for the flavonoids were as aglycones. The results of this study for the content of selected flavonoids (apigenin, luteolin, kaempferol, quercetin and isorhamnetin) in 100 edible vegetables and fruits were shown in Table 2. Since myricetin was sensitive to acid hydrolysis and rarely occurred in vegetables (Huang *et al.*, 2007), we did not quantify it in the samples using the acid extraction method. The results were listed as means and standard deviation.

It was the first time that the flavonoid contents of near half of the plant species analysed here were reported. Only 53 samples in this study have previously been reported (measured as raw vegetables or fruits) in the USDA flavonoid database (i). Flavonoid content is known to be highly dependent on the cultivar. So data in this study provided additional information as comprehensive a listing as possible to the USDA flavonoid database.

Comparison to literature values The flavonoid contents of celery and parsley were also reported in the USDA database (i). Similar to our results, apigenin was the dominant type of flavonoid in celery. However, the mean value of apigenin (1.70-19.10 mg/100 g) reported in the USDA

Table 2–1. Content of quercetin,kaempferol,isorhamnetin,apigenin and luteolin among 100 edible vegetables and fruits consumed in Harbin at the time of their most frequent consumption. (mg kg⁻¹ FW, $n=3$, mean \pm SD)

Food Description	Season	Qu	Ka	Is	Lu	Ap	TF
White radish, without peel (<i>Raphanus sativus</i> Linn.)	Winter	5.2 \pm 0.7	32.3 \pm 4.4	—	19.5 \pm 2.7	2.2 \pm 0.3	59.2
Carrot, without peel (<i>Daucus carota</i> Linn. var. <i>sativa</i> Hoffm.)	Winter	9.3 \pm 1.3	5.2 \pm 0.7	41.0 \pm 5.7	22.1 \pm 2.6	—	77.6
Lettuce stem, without peel (<i>Lactuca sativa</i> Linn.)	Autumn	—	—	—	—	—	—
Green radish, without peel (<i>Raphanus sativus</i> Linn.)	Autumn	3.5 \pm 0.0	40.7 \pm 3.9	—	—	—	44.2
Red radish, without peel (<i>Raphanus sativus</i> Linn.)	Autumn	—	—	—	1.5 \pm 0.2	—	1.5
Radish, red flesh, without peel (<i>Raphanus sativus</i> Linn.)	Winter	—	3.7 \pm 0.1	—	—	—	3.7
Radish, small, red skin (<i>Raphanus sativus</i> Linn.)	Spring	—	10.4 \pm 0.2	5.8 \pm 0.2	8.2 \pm 0.2	—	24.4
Hyacinth bean, green (<i>Lablab purpureus</i> (Linn.) Sweet)	Autumn	7.4 \pm 0.2	5.6 \pm 0.4	29.9 \pm 0.5	32.2 \pm 1.3	15.3 \pm 0.6	90.4
Holland bean (<i>Pisum sativum</i> Linn.)	Spring	24.6 \pm 0.2	21.3 \pm 0.1	44.0 \pm 2.6	14.2 \pm 0.5	6.0 \pm 0.1	110.1
Pea (<i>Pisum sativum</i> Linn.)	Spring	—	—	—	2.5 \pm 0.8	—	2.5
Soybean sprouts (<i>Glycine max</i> (L.) Merr.)	Spring	—	46.9 \pm 1.3	—	—	—	46.9
Mung bean sprouts (<i>Phaseolus minimus</i>)	Summer	—	5.0 \pm 0.4	—	2.2 \pm 0.3	5.2 \pm 0.4	12.4
Black-eyed pea (<i>Vigna unguiculata sinensis</i>)	Summer	1.5 \pm 0.0	21.1 \pm 0.5	10.0 \pm 0.3	6.9 \pm 0.6	2.9 \pm 0.2	42.4
Soybean (<i>Glycine max</i> (L.) Merr.)	Autumn	1.7 \pm 0.0	—	—	9.4 \pm 1.2	—	11.1
Kidney bean, green (<i>Phaseolus vulgaris</i> L.)	Autumn	3.7 \pm 0.0	10.0 \pm 0.6	26.6 \pm 0.5	23.6 \pm 0.7	10.8 \pm 0.3	74.7
Pea greens (<i>Pisum sativum</i> Linn.)	Spring	1.8 \pm 0.1	62.8 \pm 2.2	—	2.8 \pm 0.1	2.5 \pm 0.2	69.9
Eggplant,long,pale green skin, without peel (<i>Solanum incanum</i> L.)	Spring	4.3 \pm 0.2	17.5 \pm 0.4	14.6 \pm 0.3	19.1 \pm 0.5	8.6 \pm 0.3	64.1
Eggplant, small, dark purple skin, without peel (<i>Solanum incanum</i> L.)	Spring	4.1 \pm 0.3	21.1 \pm 1.5	27.5 \pm 2.0	22.0 \pm 1.5	7.8 \pm 0.6	82.5
Eggplant, long, dark purple skin, without peel (<i>Solanum incanum</i> L.)	Summer	—	—	—	—	—	—
Tomato (<i>Lycopersicon esculentum</i> Miller)	Autumn	6.7 \pm 0.3	12.3 \pm 0.4	31.2 \pm 0.7	30.7 \pm 0.7	24.2 \pm 1.0	105.1
Green pepper (<i>Capsicum annum</i> Linn.)	Summer	—	—	—	2.2 \pm 0.0	—	2.2

Table 2–2. (Continued)

Food Description	Season	Qu	Ka	Is	Lu	Ap	TF
Sweet peper, green (<i>Capsicum annuum</i> Linn. var. <i>grossum</i> (L.) Sendt.)	Summer	—	—	—	2.6±0.1	—	2.6
Hot pepper, green (<i>Capsicum annuum</i> Linn.)	Summer	1.6±0.2	—	—	25.4±0.5	5.0±0.0	32.0
Chinese wax gourd, without peel (<i>Benincasa hispida</i> (Thunb.) Cogn.)	Summer	2.5±0.1	9.8±0.6	10.2±1.1	17.7±1.1	4.0±0.3	44.2
Cucumber (<i>Cucumis sativus</i> Linn.)	Summer	2.2±0.1	9.0±0.5	10.7±0.6	12.4±0.6	4.7±0.3	39.0
Balsampear (<i>Momordica charantia</i> Linn.)	Summer	—	—	—	2.1±0.2	—	2.1
Pumpkin, without peel (<i>Cucurbita moschata</i> (Duch. ex Lam.) Duch. ex Poiret)	Autumn	3.8±0.3	22.6±1.8	34.8±2.8	18.1±1.5	8.8±0.8	88.1
Loofah, without peel (<i>Luffa cylindrica</i> (Linn.) Roem.)	Spring	—	13.4±1.6	15.7±0.7	14.3±1.1	8.0±0.6	51.4
Zucchini, green (<i>Cucurbita pepo</i> Linn.)	Summer	—	—	15.1±0.2	18.8±0.0	5.3±0.1	39.2
Onion, red skin (<i>Allium cepa</i> L.)	Autumn	10.5±1.4	48.2±2.2	—	—	34.5±0.2	93.2
Onion, white skin (<i>Allium cepa</i> L.)	Autumn	14.2±0.6	41.3±2.4	—	—	26.2±1.2	81.7
Leek (<i>Allium porrum</i> L.)	Summer	9.3±0.2	117.8±2.8	—	33.1±0.9	—	160.2
Garlic (<i>Allium sativum</i> Linn.)	Summer	85.0±12.1	—	—	—	—	85.0
Leek flower (<i>Allium porrum</i> L.)	Summer	13.5±1.2	—	27.6±1.2	24.5±0.8	—	65.6
Chive (<i>Allium sativum</i> Linn.)	Spring	4.0±0.5	59.0±1.8	19.1±1.9	21.7±0.4	—	103.8
Garlic stalk (<i>Allium sativum</i> Linn.)	Spring	32.7±0.3	—	—	106.7±0.9	81.7±1.5	221.1
Garlic flowering stalk (<i>Allium sativum</i> Linn.)	Spring	2.4±0.3	—	14.9±0.6	10.3±2.2	—	27.6
Chievs flowering stalk (<i>Allium sativum</i> Linn.)	Spring	—	7.5±0.4	—	7.5±0.4	—	15.0
Welsh onion (<i>Allium fistulosum</i> Linn.)	Winter	32.2±1.8	—	—	18.7±1.6	6.8±0.8	57.7
Shallot (<i>Allium fistulosum</i> Linn.)	Summer	3.4±0.1	116.7±4.3	5.1±0.3	20.0±0.5	—	145.2
Chinese cabbage (<i>Brassica pekinensis</i> (Lour.) Rupr.)	Winter	—	32.7±5.3	46.4±4.4	—	—	79.1
Chinese pakchoi (<i>Brassica pekinensis</i> (Lour.) Rupr.)	Summer	—	39.9±6.9	—	22.2±3.7	—	62.1
Rape (<i>Brassica campestris</i> L.)	Summer	—	58.9±0.8	6.2±0.3	—	—	65.1
Cauliflower (<i>Brassica oleracea</i> Linnaeus var. <i>botrytis</i> Linnaeus)	Autumn	2.3±0.0	7.4±0.2	40.0±0.5	13.1±0.2	5.1±0.1	67.9
Spinach (<i>Spinacia oleracea</i> L.)	Summer	—	8.9±0.4	—	—	—	8.9
Celery leaf (<i>Apium graveolens</i> Linn.)	Summer	—	4.6±0.3	—	23.1±1.1	139.3±5.2	167.0
Celery stalk (<i>Apium graveolens</i> Linn.)	Summer	—	7.7±0.3	—	17.4±7.1	8.4±0.4	33.5
Parsley (<i>Apium graveolens</i> Linn.)	Winter	5.0±0.1	18.5±0.3	11.2±1.0	14.2±0.3	4.4±0.1	53.3
Water spinach (<i>Lpomoa aquatica</i> Forsk)	Summer	3.2±0.1	8.1±0.3	—	—	—	11.3
Endive (<i>Sonchus oleraceus</i> Linn.)	Summer	30.6±1.6	—	7.2±0.3	8.9±0.4	—	46.7
Lettuce, green (<i>Lactuca sativa</i> Linn. var. <i>ramosa</i> Hort.)	Summer	32.2±1.0	—	12.3±0.5	2.1±0.0	—	46.6
Lettuce, red (<i>Lactuca sativa</i> Linn. var. <i>ramosa</i> Hort.)	Summer	106.5±3.0	23.6±3.4	3.2±0.2	57.2±2.6	—	190.5
Coriander (<i>Coriandrum sativum</i> Linn.)	Summer	19.2±1.1	—	12.8±0.6	—	—	32.0
Romaine lettuce (<i>Chicorium endiva</i> L.)	Summer	27.2±0.1	—	14.4±0.4	6.4±0.0	—	48.0
Chrysanthemum crowndaisy (<i>Chrysanthemum coronarium</i> Linn.)	Summer	4.5±0.2	5.5±0.3	—	26.0±1.0	—	36.0
Fennel (<i>Foeniculum vulgare</i> Mill.)	Summer	3.7±0.0	7.4±0.3	—	29.5±0.4	5.2±0.3	45.8
Potherb mustard (<i>Brassica juncea</i> (Linnaeus) Czernajew)	Autumn	29.3±1.3	482.0±21.0	333.6±14.4	—	8.1±0.0	853.0
Radish leaf, red root, small (<i>Raphanus sativus</i> Linn.)	Spring	—	58.7±3.5	277.4±15.8	179.5±10.6	—	515.6
Kale, green (<i>Brassica oleracea</i> Linnaeus var. <i>capitata</i> Linnaeus)	Summer	4.8±0.3	24.0±2.3	59.8±4.1	23.9±2.0	2.8±0.2	115.3
Cabbage, green (<i>Brassica oleracea</i> L. cv. <i>Alba</i> DC.)	Spring	4.9±0.0	31.2±0.2	—	32.7±0.2	—	68.8
Kale, purple (<i>Brassica oleracea</i> Linnaeus var. <i>capitata</i> Linnaeus)	Summer	4.3±0.1	51.4±0.9	104.4±1.2	12.0±0.1	2.6±0.1	174.7
Broccoli (<i>Brassica oleracea</i> Linnaeus var. <i>botrytis</i> Linnaeus)	Summer	5.3±0.3	21.1±0.6	—	—	—	26.4
Amaranth, purple (<i>Amaranthus cruentus</i> L.)	Summer	—	38.7±1.0	—	—	—	38.7
Lotus root, without peel (<i>Nelumbo nucifera</i> Gaertn.)	Spring	3.9±0.7	—	—	—	—	3.9
Potato, white, without peel (<i>Solanum tuberosum</i> L.)	Winter	4.3±0.2	46.1±5.6	19.1±1.2	26.7±1.2	—	96.2
Sweet potato, white flesh, without peel (<i>Ipomoea batatas</i> (L.) Lam.)	Winter	23.8±2.5	—	—	55.4±4.0	—	79.2
Yam, without peel (<i>Dioscorea zingiberensis</i> C. H. Wright)	Spring	4.5±0.0	34.3±0.2	34.1±0.5	—	8.7±0.2	81.6
Ginger, with peel (<i>Zingiber officinale</i> Roscoe)	Autumn	—	315.0±46.0	—	10.2±2.1	—	325.2
Taro, without peel (<i>Colocasia esculentum</i> (L.) Schott)	Autumn	—	—	—	—	—	—
Toona leaf (<i>Toona sinensis</i> (A. Juss.) Roem.)	Spring	379.7±19.7	417.3±22.9	—	—	8.7±0.7	805.7
Toona sprout (<i>Toona sinensis</i> (A. Juss.) Roem.)	Spring	71.3±2.3	45.5±6.7	—	—	—	116.8
Acanthopanax leaf (<i>Acanthopanax senticosus</i> (Rupr. et Maxim.) Harms)	Spring	6.6±0.5	5.6±0.5	—	—	—	12.2
Red fuji apple, without peel (<i>Malus pumila</i> Mill.)	Autumn	31.5±0.3	108.3±1.0	—	—	—	139.8
Apple, red delicious, without peel (<i>Malus pumila</i> Mill.)	Autumn	—	50.7±7.1	144.2±9.7	149.5±4.5	—	344.4
Apple, gala, without peel (<i>Malus pumila</i> Mill.)	Autumn	51.6±3.2	113.8±7.5	—	135.9±8.3	—	301.3
Pear, green, without peel (<i>Pyrus communis</i> Linn.)	Autumn	2.4±0.1	12.9±0.7	6.8±0.4	4.3±0.2	—	26.4
Pear, white, without peel (<i>Pyrus communis</i> Linn.)	Autumn	3.7±0.2	—	—	—	—	3.7

Table 2–3. (Continued)

Food Description	Season	Qu	Ka	Is	Lu	Ap	TF
Pear, yellow, without peel (<i>Pyrus communis</i> Linn.)	Autumn	20.0±0.9	—	—	—	—	20.0
Plum (<i>Prunus</i> spp.)	Summer	3.4±0.6	31.7±1.2	52.3±3.0	39.8±0.4	—	127.2
Apricot (<i>Armeniaca vulgaris</i> Lam.)	Spring	—	—	—	—	2.3±0.1	2.3
Peach (<i>Prunus persica</i> batsch)	Summer	—	14.3±1.7	—	33.9±4.2	—	48.2
Date (<i>Ziziphus jujuba</i> Mill.)	Autumn	5.1±0.3	44.0±2.5	113.7±7.8	48.8±3.6	41.0±2.7	252.6
Hawthorn (<i>Crataegus pinnatifida</i> Bge.)	Autumn	2.9±0.1	9.3±0.4	18.5±0.8	8.3±0.3	10.7±0.3	49.7
Nectarine (<i>Prunus persica</i> var. <i>nucipersica</i>)	Summer	11.9±0.8	49.6±3.9	—	49.7±3.1	—	111.2
Strawberry (<i>Fragaria X ananassa</i> Duch.)	Spring	—	2.8±0.1	5.7±0.1	—	2.4±0.1	10.9
Grape, small, green, with peel (<i>Vitis vinifera</i> Linn.)	Autumn	11.9±0.3	89.1±4.0	—	—	—	101.0
Rose fragrant grape, rose flavored, without peel (<i>Vitis vinifera</i> Linn.)	Autumn	—	—	—	—	—	—
Giant grape, green, without peel (<i>Vitis vinifera</i> Linn.)	Autumn	28.5±0.1	—	—	—	—	28.5
Persimmon, without peel (<i>Diospyros vaccinioides</i> Lindl.)	Autumn	—	—	12.3±0.8	8.1±0.6	8.1±0.4	28.5
Orange (<i>Citrus reticulata</i> Blanco cv. <i>Tankan</i>)	Winter	1.7±0.2	5.1±0.5	8.7±0.8	4.5±0.4	—	20.0
Mandarin (<i>Fortunella margarita</i> (Lour.) Swingle)	Autumn	8.6±1.2	64.2±9.3	44.9±3.6	37.8±7.1	—	155.5
Shaddock (<i>Citrus grandis</i> (Linn.) Osbeck)	Winter	10.9±0.3	—	—	—	—	10.9
Orange, navel (<i>Citrus sinensis</i> (L.) Osbeck)	Winter	19.6±1.2	—	96.7±3.8	—	—	116.3
Kiwi fruit (<i>Actinidia arguta</i> var. <i>giraldii</i>)	Winter	—	11.1±0.6	6.0±0.3	3.9±0.2	—	21.0
Pineapple (<i>Ananas comosus</i> (Linn.) Merr.)	Spring	—	—	—	66.9±7.0	—	66.9
Banana (<i>Musa nana</i> Lour.)	Winter	—	3.1±0.3	8.1±0.7	5.7±0.5	—	16.9
Watermelon (<i>Citrullus lanatus</i> (Thunb.) Matsum. et Nakai)	Autumn	6.7±0.2	13.9±1.3	—	70.6±2.3	—	91.2
Muskmelon, without peel (<i>Cucumis melo</i> Linn.)	Summer	13.7±1.3	—	—	—	—	13.7
Tomato, cherry, yellow (<i>Lycopersicon esculentum</i> var. <i>cerasiforme</i>)	Summer	7.6±0.1	21.5±0.8	—	19.1±0.2	19.9±0.3	68.1
Tomato, cherry, red (<i>Lycopersicon esculentum</i> var. <i>cerasiforme</i>)	Summer	—	8.3±0.4	—	48.6±0.7	20.8±0.4	77.7

Qu: quercetin; Ka: kaempferol; Is: isorhamnetin; Ap: apigenin; Lu: luteolin; TF: total flavonoids (quercetin, kaempferol, isorhamnetin, apigenin and luteolin).

“—”: not detected (below the detection limit).

database (i) was lower than that of our results (139.3 ± 5.2 mg kg⁻¹), and the value previously reported (2.5 mg/100 g) (Ray-Yu, Y., Shou, L. and George, K. 2008) was also lower than that of our results (139.3 ± 5.2 mg kg⁻¹). Different from our results, apigenin was the dominant type of flavonoid in parsley and a higher value (226 mg/100 g) was reported in the USDA database (i). In our study, the level of apigenin in parsley was only 4.4 ± 0.1 mg kg⁻¹, lower than the content of luteolin, kaempferol, quercetin and isorhamnetin in parsley. From Table 2, we also found that the variation in the composition and content of five flavonoids between celery and parsley is very large. For celery, we investigated the level of five flavonoids in celery leaf and celery stalk, respectively. Except for apigenin, the composition and levels of other four flavonoids were similar between celery leaf and celery stalk. The higher apigenin content of celery leaf (139.3 ± 5.2 mg kg⁻¹) relative to celery stalk (8.4 ± 0.4 mg kg⁻¹) can be explained by the previous report that flavonoids occur predominately in the leaves (Herrmann, 1976).

Two types of lettuce exhibited very wide differences in total flavonoid content, 46.6 mg kg⁻¹ for green lettuce and 190.5 mg kg⁻¹ for red lettuce. As can be seen in Table 2, the higher flavonoid level of red lettuce must be due to a much higher quercetin content (106.5 mg kg⁻¹). The higher flavo-

noid level of red lettuce compared with green lettuce has already been reported by other authors (Dupont *et al.*, 2000; Paola *et al.*, 2004). The mean luteolin content of lettuce was of 29.7 mg kg⁻¹ in our study, these results are similar to those of Paola *et al.* (2004), 2 mg/100 g of FW. Different from Paola *et al.* (2004), we detected isorhamnetin in lettuce and the mean isorhamnetin level was of 7.8 mg kg⁻¹. According to Dupont *et al.* (2000) variations within a variety could be due to different agronomic conditions, tissue type (red to green), and leaves sampled (outer or inner).

In addition, none of the flavonoids investigated could be detected in lettuce stem (without peel), eggplant (long, dark purple skin, without peel), taro (without peel) and rose fragrant grape (rose flavored, without peel). As we all known that flavonoids are often produced in direct response to environmental conditions. It has been documented that the flavonoid content is dependent on ultraviolet light and CO₂ levels (Caldwell *et al.*, 2005; Daniel *et al.*, 1999). Therefore it is not surprising that there are differences in the flavonoid levels for similar food collected from different regions at different time.

Food sources of selected flavonoids In this study, high levels of apigenin, luteolin, kaempferol, quercetin and isorhamnetin were listed in Table 3. It was noticed that most of

leafy vegetables are rich in flavonols and flavones. Potherb mustard, lettuce (red), toona leaf, celery leaf, garlic, garlic stalk, apple, date, ginger (with peel) and radish leaf (red root, small) may be considered as the main food sources of flavonols and flavones in China. Whether these vegetables and apples may be considered as the main food sources of the dietary flavonol intake and dietary flavone intake in China, it must be to do a population dietary survey and consider the amounts of consumption of these vegetables and apples. So far, there has not been the population dietary flavonoids intake survey in our country. Tea, onions, and apples are the most predominant food sources of flavonols in The Netherlands (Hertog *et al.*, 1993), Denmark (Justesen *et al.*, 1997), and United States (Rimm *et al.*, 1996). Variations in the dietary sources are influenced by such factors as feeding habits and cultural changes.

The major flavonoids that we found in vegetables and fruits are kaempferol, followed by luteolin and quercetin (Table 3). Among 100 samples we studied, 68 samples all contained kaempferol (above the limit of detection), and the content of kaempferol in 48 samples were ≥ 10.0 mg kg⁻¹. However, among 100 samples, 66 samples all contained quercetin (above the limit of detection), and the content of quercetin only in 25 samples were ≥ 10.0 mg kg⁻¹. This suggested that although quercetin is widely distributed in plant species, it is generally present only in low concentrations, except for specific plant foods with very high quercetin content such as garlic, cruciferous vegetables. Kumpulainen *et al.* (1999) reported quercetin presents in high concentrations in onions. In our study, low quercetin levels were detected in onions (10.5–14.2 mg kg⁻¹) (Table 2) which were obviously

lower than those of the values (7.29–33.43 mg/100 g) in the USDA database (i) and the values (38.3–93.6 mg/100 g) of Paola *et al.* (2004). The difference in the reported values may be from different cultivars and different growing conditions.

Seasonal variations It was anticipated that seasonal variations might result in differences in flavonoid levels. In our study, we investigated the effect of season on flavonoid amounts in foods cultivated in China. Table 4 reported the flavonoid content of ten vegetables consumed in Harbin at two different seasons of the year. It was observed that the total flavonoid content due to season factors was quite variable for vegetables such as kale (purple) (6.6 and 174.7 mg kg⁻¹), chrysanthemum crowndaisy (16.5 and 36.0 mg kg⁻¹), amaranth (purple) (6.9 and 38.7 mg kg⁻¹), and coriander (2.3 and 32.0 mg kg⁻¹), which presented much higher flavonoid levels during summer due to a higher incidence of light compared with autumn. This is in accordance with Hertog *et al.* (1992), who reported that variations in flavonoid levels due to seasonal factors were large in leafy vegetables, and flavonoid levels in the summer in these vegetables can be three to five times greater than in other seasons, and seasonal variability was low for other vegetables. Seasonal variability was, however, low for chinese pakchoi (53.0 and 62.1 mg kg⁻¹) in our study. Similar to our results, Hertog *et al.* (1992) did not observe seasonal variation in flavonoid content for onion (red skin). However, tomato and carrot (without peel) presented a higher flavonoid content in autumn than spring and in autumn than winter, respectively. These variabilities could be ascribed to different light exposure at different seasons.

A limitation of this study is that we did not evaluate the effect of location on flavonoid contents in vegetables and

Table 3. Content ranges of flavonoids (mg kg⁻¹ FW) and food rich in individual flavonoids.

	Qu	Ka	Is	Lu	Ap	TF
Food rich in individual flavonoids						
First	Toona leaf (<i>Toona sinensis</i> (A. Juss.) Roem.)	Potherb mustard (<i>Brassica juncea</i> (Linnaeus) Czernajew)	Potherb mustard (<i>Brassica juncea</i> (Linnaeus) Czernajew)	Radish leaf, red root, small (<i>Raphanus sativus</i> Linn.)	Celery leaf (<i>Apium graveolens</i> Linn.)	Potherb mustard (<i>Brassica juncea</i> (Linnaeus) Czernajew)
Second	Lettuce, red (<i>Lactuca sativa</i> Linn. var. <i>ramosa</i> Hort.)	Toona leaf (<i>Toona sinensis</i> (A. Juss.) Roem.)	Radish leaf, red root, small (<i>Raphanus sativus</i> Linn.)	Apple, red delicious, without peel (<i>Malus pumila</i> Mill.)	Garlic stalk (<i>Allium sativum</i> Linn.)	Toona leaf (<i>Toona sinensis</i> (A. Juss.) Roem.)
Third	Garlic (<i>Allium sativum</i> Linn.)	Ginger, with peel (<i>Zingiber officinale</i> Roscoe)	Apple, red delicious, without peel (<i>Malus pumila</i> Mill.)	Apple, gala, without peel (<i>Malus pumila</i> Mill.)	Date (<i>Ziziphus jujuba</i> Mill.)	Radish leaf, red root, small (<i>Raphanus sativus</i> Linn.)
Content ≥ 10.0 mg kg ⁻¹						
n	25	48	35	47	11	86
Content < the detection limit						
n	34	32	55	33	63	4
Maximum (mg kg ⁻¹)	379.7 \pm 19.7	482.0 \pm 21.0	333.6 \pm 14.4	179.5 \pm 10.6	139.3 \pm 5.2	853

Qu: quercetin; Ka: kaempferol; Is: isorhamnetin; Ap: apigenin; Lu: luteolin; TF: total flavonoids (quercetin, kaempferol, isorhamnetin, apigenin and luteolin).

Table 4. The flavonoid content of ten vegetables consumed in Harbin at two different seasons of the year (mg kg⁻¹ FW, *n*=3, mean±SD).

Food Description	Season	Qu	Lu	Ka	Is	Ap	TF
Tomato (<i>Lycopersicon esculentum</i> Miller)	Spring	6.8±0.6	25.3±2.4	23.3±2.4	—	21.7±2.0	77.1
	Autumn	6.7±0.3	30.7±0.7	12.3±0.4	31.2±0.7	24.2±1.0	105.1
Onion, red skin (<i>Allium cepa</i> L.)	Autumn	10.5±1.4	—	48.2±2.2	—	34.5±0.2	93.2
	Winter	—	—	—	83.5±12.7	8.0±1.2	91.5
Carrot, without peel (<i>Daucus carota</i> Linn. var. <i>sativa</i> Hoffm.)	Autumn	20.5±1.7	84.7±6.0	88.1±5.4	133.3±9.2	40.9±2.0	367.5
	Winter	9.3±1.3	22.1±2.6	5.2±0.7	41.0±5.7	—	77.6
Kale, purple (<i>Brassica oleracea</i> Linnaeus var. <i>capitata</i> Linnaeus)	Summer	4.3±0.1	51.4±0.9	104.4±1.2	12.0±0.1	2.6±0.1	174.7
	Autumn	—	6.6±0.3	—	—	—	6.6
Chrysanthemum crowndaisy (<i>Chrysanthemum coronarium</i> Linn.)	Summer	4.5±0.2	5.5±0.3	—	26.0±1.0	—	36.0
	Autumn	10.5±0.6	—	—	6.0±0.3	—	16.5
Coriander (<i>Coriandrum sativum</i> Linn.)	Summer	19.2±1.1	—	—	12.8±0.6	—	32.0
	Autumn	2.3±0.1	—	—	—	—	2.3
Amaranth, purple (<i>Amaranthus cruentus</i> L.)	Summer	—	38.7±1.0	—	—	—	38.7
	Autumn	—	6.9±0.0	—	—	—	6.9
Leek (<i>Allium porrum</i> L.)	Spring	—	103.3±0.8	13.5±0.6	18.5±0.5	7.8±0.4	143.1
	Summer	9.3±0.2	117.8±2.8	—	33.1±0.9	—	160.2
Chinese pakchoi (<i>Brassica pekinensis</i> (Lour.) Rupr.)	Spring	4.0±0.7	—	—	45.8±0.8	3.2±0.1	53.0
	Summer	—	39.9±6.9	—	22.2±3.7	—	62.1
Celery leaf (<i>Apium graveolens</i> Linn.)	Spring	—	51.2±3.7	5.9±0.5	—	329.0±24.1	386.1
	Autumn	—	79.1±6.5	14.6±0.7	5.0±0.5	375.2±34.4	473.9

Qu: quercetin; Ka: kaempferol; Is: isorhamnetin; Ap: apigenin; Lu: luteolin; TF: total flavonoids (quercetin, kaempferol, isorhamnetin, apigenin and luteolin).

“—”: not detected (below the detection limit).

fruits because of our limited material, financial and human resources. Further investigation, in cooperation with other research institutes, is required to evaluate the effect of location on the content of flavonoids in foods cultivated in China in the future.

Conclusion

In this study the flavonoid content and composition of 100 edible vegetables and fruits frequently consumed in Harbin city of China were analyzed. These data were collected for the establishment of China's flavonoid database. With data on the content of flavonoids currently obtained in this study, the assessment of the daily flavonoid intake by Chinese population can be made. Our data thus provided a basis for epidemiological studies investigating the relationship between the flavonoid intake and cancer risk.

Acknowledgements Financial support from the Education Department of Heilongjiang Province (11541157) is gratefully acknowledged.

References

- Bosetti, C., Rossi, M., McLaughlin, J.K., Negri, E., Talamini, R., Lagiou, P., Montella, M., Ramazzotti, V., Franceschi, S. and LaVecchia, C. (2007). Flavonoids and the risk of renal cell carcinoma. *Cancer Epidemiol. Biomarkers Prev.*, **16**, 98.
- Beecher, G.R. (2003). Overview of dietary flavonoids: Nomenclature, occurrence and intake. *J. Nutr.*, **133**, 3248S-3254S.
- Caldwell, C.R., Britz, S.J. and Mirecki, R.M. (2005). Effect of temperature, elevated carbon dioxide, and drought during seed development on the isoflavone content of dwarf soybean [*Glycine max* (L.) Merrill] grown in controlled environments. *J. Agric. Food Chem.*, **53**, 1125-1129.
- Cao, J., Zhao, X.J., Wu, K., Zhang, Y. and Zhang, Y.Q. (2008). Simultaneous Determination of Five Flavonoid Compounds in Vegetables and Fruits by High Performance Liquid Chromatography. *Chinese J. Prev. Med. Inf.*, **7**, 525-527.
- Daniel, O., Meier, M.S., Schlatter, J. and Frischknecht, P. (1999). Selected phenolic compounds in cultivated plants: ecologic functions, health implications, and modulation by pesticides. *Environ. Health Perspect.*, **107**, 109-114.
- Dupont, M. S., Mondim, Z., Williamson, G. and Price, K. R. (2000). Effect of Variety, Processing, and Storage on the Flavonoid Glycoside Content and Composition of Lettuce and Endive. *J. Agric. Food Chem.*, **48**, 3957-3964.
- Hertog, M.G.L., Hollman, P.C.H., Katan, M.B. and Kromhout, D. (1993). Intake of potentially anticarcinogenic flavonoids and their determinants in adults in The Netherlands. *Nutr. Cancer*, **20**, 21-29.
- Hertog, M.G.L., Hollman, P.C.H. and Katan, M.B. (1992). Content of potentially anticarcinogenic flavonoids of 28 vegetables and 9 fruits commonly consumed in The Netherlands. *J. Agric. Food Chem.*, **40**, 2379-2383.
- Herrmann, K. (1976) Flavonols and flavones in food plants. A review. *J. Food Technol.*, **11**, 433-448.

- Huang, Z.L., Wang, B.W., Eaves, D.H., Shikany, J.M. and Pace, R.D. (2007). Phenolic compound profile of selected vegetables frequently consumed by African Americans in the southeast United States. *Food Chem.*, **103**, 1395-1402.
- Justesen, U., Knuthsen, P. and Leth, T. (1997). Determination of plant polyphenols in Danish foodstuffs by HPLC-UV and LC-MS detection. *Cancer Lett.*, **114**, 165-7.
- Kumpulainen, J.T., Lehtonen, M. and Mattila, P. (1999). Trolox equivalent antioxidant capacity of average flavonoids intake in Finland, In J. T. Kumpulainen and J. T. Salonen (eds.), *Natural Antioxidants and Anticarcinogens in Nutrition*, Proceedings of the Second International Conference on Natural Antioxidants, Cambridge, United Kingdom: The Royal Society of Chemistry, pp. 141-150.
- Kinoshita, T., Lepp, Z. and Chuman, H. (2005). Construction of a novel database for flavonoids. *J. Med. Invest.*, **52**, 291-292.
- Middleton, E., Kandaswami, C. and Theoharides, T.C. (2000). The effects of plant flavonoids on mammalian cells: Implications for inflammation, heart disease, and cancer. *Pharmacol. Rev.*, **52**, 673-751.
- Nijveldt, R.J., van Nood, E., van Hoorn, D.E.C., Boelens, P.G., van Norren, K. and van Leeuwen, P.A.M. (2001). Flavonoids: a review of probable mechanisms of action and potential applications. *Am. J. Clin. Nutr.*, **74**, 418-25.
- Paola R.A., Maria I.G. and Franco M.L. (2004). Flavonoids in Vegetable Foods Commonly Consumed in Brazil and Estimated Ingestion by the Brazilian Population. *J. Agric. Food Chem.*, **52**, 1124-1131.
- Peterson, J. and Dwyer, J. (2000). An informatics approach to flavonoid database development. *J. Food Compos. Anal.*, **13**, 441-454.
- Rimm, E.B., Katan, M.B., Ascherio, A., Stampfer, M.J. and Willett, W.C. (1996). Relation between intake of flavonoids and risk for coronary heart disease in male health professionals. *Ann. Intern. Med.*, **125**, 384-389.
- Rossi, M., Negri, E., Talamini, R., Bosetti, C., Parpinel, M., Gnagnarella, P., Franceschi, S., Dal Maso, L., Montella, M., Giacosa, A. and La Vecchia, C. (2006). Flavonoids and colorectal cancer in Italy. *Cancer Epidemiol. Biomarkers Prev.*, **15**, 1555-1558.
- Ray-Yu, Y., Shou, L. and George, K. (2008). Content and distribution of flavonoids among 91 edible plant species. *Asia Pac. J. Clin. Nutr.*, **17**(S1), 275-279.
- Scalbert, A. and Williamson, G. (2000). Dietary intake and bioavailability of polyphenols. *J. Nutr.*, **130**, 2073S-2085S.

URL cited

- i) <http://www.ars.usda.gov/nutrientdata> (Apr.27.2009)