

Flavonoid Composition of Fruit Tissues of Citrus Species

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Received August 2, 2005; Accepted September 12, 2005

An HPLC analysis was performed on the concentrations of flavonoids in 42 species and cultivars of the *Citrus* genus and those of two *Fortunella* and one *Poncirus* species according to the classification system established by Tanaka. The composition of 8 flavanones and 9 flavone/ols for these species was determined in the albedo, flavedo, segment epidermis and juice vesicle tissues, and those in the fruit and peel tissues were calculated from the composition data of the tissues. A principal component analysis showed that such neohesperidosyl flavonoids as neoeriocitrin, naringin, neohesperidin, and rhoifolin had large factor loading values in the first principal component for each tissue. The flavonoid composition of citrus fruits was approximately the same within each section of Tanaka's system, except for the species in the *Aurantium* section and those with a peculiar flavonoid composition such as Bergamot (*C. bergamia*), Marsh grapefruit (*C. paradisi*), Sour orange (*C. aurantium*), and Shunkokan (*C. shunkokan*). The *Aurantium* section included both naringin-rich and hesperidin-rich species.

Key words: *Citrus*; flavanone; flavone; flavonol; taxonomy

Citrus plants contain a wide range of flavonoid constituents, some of which, *e.g.*, hesperidin, naringin, and polymethoxylated flavones (PMFs), are characteristic of them and others such as rutin and quercetin are common in the plant kingdom. Attempts have recently been made to find biological activities among citrus flavonoids. For instance, naringin has been found to lower the total cholesterol and low-density lipoprotein

cholesterol levels in plasma,¹⁾ while the administration of hesperetin and its metabolites significantly lowered the total cholesterol and triglyceride concentrations in plasma.²⁾ Hesperidin and diosmin, both alone and in combination, act as a chemopreventive agent against colon carcinogenesis induced by azoxymethane.³⁾ The polymethoxylated flavone, nobiletin, has been reported to effectively down-regulate the production of pro-matrix metalloproteinase and to interfere with the proliferation of synovial fibroblasts.⁴⁾ Tangeretin has been reported to have a suppressive effect on malignant tumor invasion and metastasis.⁵⁾ The purpose of these present studies is to prevent chronic diseases through the daily intake of citrus fruits and to increase their added value.

Paper chromatography, thin-layer chromatography, and open column chromatography have all been used to analyze citrus flavonoids.^{6–9)} These methods are effective for the separation and identification of flavonoids, but of limited quantitative value. The introduction of the HPLC technique has led to better resolution and more precise quantification.^{10–13)} Past studies have attempted to apply flavonoid composition to the classification and discrimination of *Citrus* species, although HPLC determinations are limited. For instance, Gaydou *et al.* have quantified the polymethoxylated flavones in mandarins (*C. reticulata*) and oranges (*C. sinensis*).¹⁴⁾ Mouly *et al.* have effectively differentiated lemon, lime, grapefruit, and sweet orange by a factorial discrimination analysis of the flavanone glycoside composition in the juice.¹⁵⁾ Kawai *et al.* have quantified 25 flavonoids in hand-squeezed juice from 34 citrus samples and analyzed the data by a principal component analysis, indicating the

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Abbreviations: ERC, eriocitrin; NER, neoeriocitrin; NRT, narirutin; NRG, naringin; HSP, hesperidin; NHP, neohesperidin; NPO, neoponcirin; PON, poncirin; RTN, rutin; IRF, isorhoifolin; RFN, rhoifolin; DSM, diosmin; NDM, neodiosmin; SNT, sinensetin; NOB, nobiletin; TNG, tangeretin; HPM, heptamethoxyflavone

peculiarities of King (*C. nobilis*) and Bergamot (*C. bergamia*).¹⁶⁾ These studies should contribute to the chemical taxonomy of *Citrus* species, although an objective extraction method is needed for this purpose, because the distribution of flavonoids in citrus tissues varies with the structure and so does the preferable extraction method.¹⁷⁾ The development of DNA marker techniques has enabled RAPD,¹⁸⁾ ISSR,¹⁹⁾ and RFLP²⁰⁾ analyses to be conducted on the taxonomy and genealogy of *Citrus* species. Concomitantly, studies to discriminate species and cultivars by ISSR²¹⁾ and CAPS²²⁾ analyses have been performed. These methods exhibit sufficient discriminative ability provided that the samples are genetically similar, but when the genetic background is not known, it takes considerable time and effort to discover effective markers.

The detailed flavonoid composition in fruit tissues of the *Citrus* species was analyzed in this study to provide data that would enable more effective utilization of flavonoid constituents and to examine the agreement of their flavonoid composition with Tanaka's classification system.

Materials and Methods

Plant materials. The citrus fruits investigated were grown and harvested at the Department of Citrus Research (National Institute of Fruit Tree Science, Okitsu, Japan). From the same tree, 5–15 mature fruits were harvested in 2003. Four different tissue samples, the flavedo, albedo, segment epidermis, and juice vesicle, were dissected, weighed, and stored at -80°C prior to their preparation.

Sample preparation. The samples were prepared and analyzed as described previously.¹³⁾ The flavedo, albedo, and segment epidermis samples were lyophilized and ground with an Ultra Centrifugal Mill (Mitamura Riken Kogyo, Tokyo, Japan) filled with a 0.5-mm filter. Portions (100 mg) of the powdered tissues were extracted for 12 h with 5 ml of methanol–DMSO (1:1, v/v) at ambient temperature. After centrifugation at 3,000 g for 10 min, the extract was decanted, and the remaining residue was extracted twice more with 1 ml of the same solution. The combined extract was diluted tenfold with distilled water and passed through a SepPak C₁₈ (0.36 g) cartridge which had been preconditioned with 5 ml of methanol and then 10 ml of 10% methanol. The cartridge was washed with 10% methanol, and the flavonoids were eluted with methanol–DMSO (1:1, v/v). The volume of the eluate was adjusted to 5 ml and filtered through a membrane filter (0.5 mm) prior to injection. The juice sample was prepared by homogenizing the juice vesicles with a mixer. The homogenate was centrifuged at 15,000 g for 20 min, and 3 ml of the resulting supernatant was passed through the SepPak cartridge as already described.

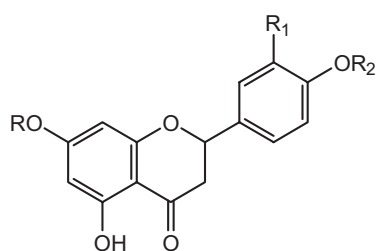
Flavonoid analysis. The structures of flavonoids investigated are presented in Fig. 1. NDM was donated by the Fruit and Vegetable Chemistry Laboratory (US Department of Agriculture, Agricultural Research Service, Pasadena, CA, U.S.A.). Other HPLC-grade flavonoid standards were purchased from Funakoshi (Tokyo, Japan), all other chemicals being of analytical grade from Wako (Osaka, Japan). The HPLC system consisted of two pumps, an autosampler, a photodiode array detector (Hitachi, Tokyo, Japan), a column oven (Shimadzu, Kyoto, Japan), and a C₁₈ RP column (LiChrospher 100 RP-18, 250 × 4.0 mm-i.d.; Merck, Darmstadt, Germany) with a cartridge guard column. The detector was monitored at 285 nm and measured spectra from 200 to 360 nm. A two-solvent gradient system of aqueous 10 mM phosphoric acid (A) and methanol (B) was used. The gradient program consisted of three periods: (1) 0–55 min, 70–55% A, (2) 55–95 min, 55–0% A, (3) 95–100 min, 100% B. The resulting chromatographic data was integrated up to 90 min. The flow-rate was 0.6 ml/min, the column was operated at 40 °C, and the sample injection volume was 10 µl. The flavonoids were identified by comparing their retention times and UV spectra with those of authentic standards stored in a data processor. The concentration of each flavonoid was calculated from the integrated peak area of the sample and the corresponding standard.

Principal component analysis. The concentrations of 17 flavonoid constituents in the citrus fruits were used as variables. The Statistica ver. 4.5 statistical analysis program (StatSoft Inc., Tulsa, OK, U.S.A.) was used to calculate and plot data from the principal component analysis. Each principal component was calculated by taking a linear combination of an eigenvector of the correlation matrix with original variables. A dendrogram was calculated by using the UPGMA (unweighted pair-group method using arithmetic averages) method based on the standardized data of flavonoid composition in *Citrus* fruits.

Results and Discussion

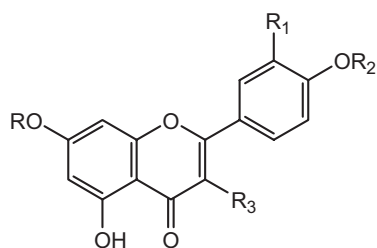
Distribution of flavonoids in citrus fruits and tissues

The species investigated are listed in Table 1 according to the morphological system established by Tanaka who classified the *Citrus* genus into 2 subgenera, 8 sections and 16 subsections, involving 149 species, in 1969.²³⁾ Swingle, an advocator of another system, classified *Citrus* into 2 subgenera and 16 species including 8 varieties in 1948.²⁴⁾ The major difference between these two systems is in how mandarins (group VII in Table 1) were treated: Swingle placed all mandarins except *C. tachibana* and *C. indica* in *C. reticulata*, whereas Tanaka separated mandarins into 36 species. Tables 2–7 show the flavonoid concentrations in each tissue of citrus fruit on a fresh weight basis. The composition data for each fruit (Table 2) were



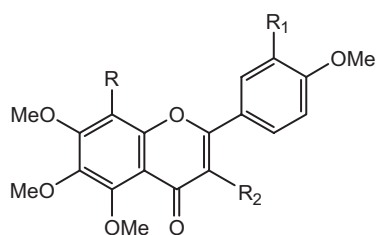
Flavanone

Eriocitrin (ERC) :	R=rutinose, R ₁ =OH, R ₂ =H
Neeriocitrin (NER):	R=neohesperidose, R ₁ =OH, R ₂ =H
Narirutin (NRT):	R=rutinose, R ₁ =R ₂ =H
Naringin (NRG):	R=neohesperidose, R ₁ =R ₂ =H
Hesperidin (HSP):	R=rutinose, R ₁ =OH, R ₂ =Me
Neohesperidin (NHP):	R=neohesperidose, R ₁ =OH, R ₂ =Me
Neoponcirin (NPO):	R=rutinose, R ₁ =H, R ₂ =Me
Poncirin (PON):	R=neohesperidose, R ₁ =H, R ₂ =Me



Flavone

Rutin (RTN):	R=H, R ₁ =OH, R ₂ =H, R ₃ =O-rutinose
Isorhoifolin (IRF):	R=rutinose, R ₁ =R ₂ =R ₃ =H
Rhoifolin (RFN):	R=neohesperidose, R ₁ =R ₂ =R ₃ =H
Diosmin (DSM):	R=rutinose, R ₁ =OH, R ₂ =Me, R ₃ =H
Neodiosmin (NDM)	R=neohesperidose, R ₁ =OH, R ₂ =Me, R ₃ =H



Polymethoxylated flavone

Sinensetin (SNT):	R=H, R ₁ =OMe, R ₂ =H
Nobiletin (NOB):	R=R ₁ =OMe, R ₂ =H
Tangeretin (TNG):	R=OMe, R ₁ =R ₂ =H
Heptamethoxyflavone (HPM):	R=R ₁ =R ₂ =OMe

Fig. 1. Structures of the Flavonoids Investigated.

calculated from those of the flavedo, albedo, segment epidermis, and juice vesicle tissues, and that for the peel tissue (Table 3) was from those of the flavedo and albedo tissues. ERC widely occurred in the species of each section excepting *Fortunella* (102) and *Poncirus* (103). Tahiti lime (#3), Eureka lemon (#7), Lumie (#9), and Koji (#41) contained ERC at the highest concentrations among the 45 species (Table 2). The tissue containing the highest concentration of this compound was flavedo of the Tahiti lime (#3) and albedo of the Eureka lemon (#7), Lumie (#9), and Koji (#41) (Tables 5 and 6). ERC was also present at high levels in the juice vesicle of these fruits (Table 4). Bergamot (#4) and Sour orange (#17) fruits contained NER in exceptionally large amounts (Table 2). In the *Limonellus* (II) section, Bergamot (#4) had a peculiar flavonoid composition in that it had large amounts of NER and NHP, but little HSP. The occurrence of NER was relatively limited to certain species. NRT occurred in all species investigated except Cabuyao (#1), Citron (#6) and Shaten yu (#11) (Table 2). The species belonging to the *Aurantium* (V) section such as Sanbokan (#16) and

Shunkokan (#22), and those to *Fortunella* (102) contained NRT in large amounts (Table 2). This flavonoid was abundant in the flavedo and segment epidermis, but scarce in the albedo of the *Citrus* species (I–VIII) (Tables 5–7). In the *Fortunella* (102) species, NRT was present at higher levels in the segment epidermis and juice vesicle than in the peel (Tables 3, 4 and 7). NRG was present in large amounts in species of the *Cephaecitrus* (IV), *Aurantium* (#15–17), and *Osmocitrus* (VI) sections, and in Bergamot (#4), Shikikitsu (#42), and Trifoliate orange (#45) (Table 2). Swingle defined Shikikitsu (#42) as a hybrid of *C. reticulata*, although it had a similar flavonoid composition not to mandarins but to the *Osmocitrus* (VI) section species.²⁵⁾ Marsh grapefruit (#12) contained the highest concentration of NRG, its respective content in the fruit tissues being 2700, 1270, 1130, and 994 mg/100 g in albedo, juice vesicle, segment epidermis, and flavedo (Tables 4–7). HSP was distributed in all fruits except part of the *Cephaecitrus* species (#10–12), Sour orange (#17), and Trifoliate orange (#45) (Table 2). It was present at a high level in fruits of the Tahiti lime (#3), Biroro (#5),

Table 1. Classification of Citrus Plants Investigated for an Analysis of Flavonoid Content^a

Specimen	Tanaka's no.	Ref.	Common name	Scientific name
<i>Citrus-Archicidrus</i>				
<i>Papeda</i>				
<i>Acutifolia</i>	I-1-1	1	Cabuyao	<i>C. macroptera</i>
<i>Obtusifolia</i>		— ^b		
<i>Longipetiolata</i>		—		
<i>Limonellus</i>				
<i>Eulimonellus</i>	II-4-13	2	Mexican lime	<i>C. aurantifolia</i>
	II-4-14	3	Tahiti lime	<i>C. latifolia</i>
<i>Megacarpa</i>	II-5-17	4	Bergamot	<i>C. bergamia</i>
<i>Pseudopapeda</i>	II-6-29	5	Biroro	<i>C. montana</i>
<i>Citrophorum</i>				
<i>Citroides</i>	III-7-31	6	Citron	<i>C. medica</i>
<i>Limonioides</i>	III-8-36	7	Eureka lemon	<i>C. limon</i>
	III-8-38	8	Sweet lemon	<i>C. limetta</i>
<i>Decumanoides</i>	III-9-48	9	Lumie	<i>C. lumia</i>
<i>Cephacitrus</i>				
<i>Decumana</i>	IV-10-56	10	Hirado buntan	<i>C. grandis</i> cv. Hirado
	IV-10-61	11	Shaten yu	<i>C. grandis</i> cv. Shytian you
<i>Intermedia</i>				
<i>Flavicarpa</i>	IV-11-62	12	Marsh grapefruit	<i>C. paradisi</i>
	IV-11-63	13	Kinukawa	<i>C. glaberrima</i>
<i>Aureocarpa</i>	IV-12-74	14	Hassaku	<i>C. hassaku</i>
<i>Aurantium</i>				
<i>Medioglobosa</i>	V-13-78	15	Natsudaidai	<i>C. natsudaidai</i>
	V-13-84	16	Sanbokan	<i>C. sulcata</i>
<i>Aurantioides</i>				
<i>Racemosa</i>	V-14-93	17	Sour orange	<i>C. aurantium</i>
<i>Contracta</i>		—		
<i>Sinenoides</i>	V-16-100	18	Valencia	<i>C. sinensis</i> cv. Valencia
	V-16-100	19	Morita navel	<i>C. sinensis</i> var <i>Brasilensis</i> cv. Morita
	V-16-105	20	Iyo	<i>C. iyo</i>
<i>Osmocitroides</i>				
<i>Temnicarpa</i>	V-17-107	21	Hyuganatsu	<i>C. tamurana</i>
<i>Compacta</i>		—		
<i>Paranobilis</i>	V-19-111	22	Shunkokan	<i>C. shunkokan</i>
<i>Citrus-Metacitrus</i>				
<i>Osmocitrus</i>				
<i>Protosmocitrus</i>		—		
<i>Euosmocitroides</i>	VI-21-113	23	Yuzu	<i>C. junos</i>
	VI-21-115	24	Sudachi	<i>C. sudachi</i>
	VI-21-121	25	Kabosu	<i>C. sphaerocarpa</i>
<i>Pseudoacrumen</i>		—		
<i>Acrumen</i>				
<i>Euacrumen</i>	VII-23-123	26	King	<i>C. nobilis</i> var <i>Knep</i>
	VII-23-124	27	Satsurna	<i>C. unshiu</i>
	VII-23-125	28	Yatsushiro	<i>C. yatsusiro</i>
<i>Microacrumen</i>				
<i>Anisodora</i>	VII-24-126	29	Keraji	<i>C. keraji</i>
	VII-24-127	30	Oto	<i>C. oto</i>
<i>Citroidora</i>	VII-25-130	31	Ponkan	<i>C. reticulata</i>
<i>Megacarpa</i>	VII-25-133	32	Dancy tangerine	<i>C. tangerina</i>
	VII-25-134	33	Clementine	<i>C. clementina</i>
	VII-25-136	34	Jimikan	<i>C. succosa</i>
	VII-25-140	35	Shikaikan	<i>C. suhuiensis</i>
<i>Microcarpa</i>				
<i>Angustifolia</i>	VII-26-143	36	Tachibana	<i>C. tachibana</i>
	VII-26-144	37	Kobenimikan	<i>C. erythroa</i>
	VII-26-145	38	Kishu	<i>C. kinokuni</i>
<i>Latifolia</i>	VII-27-148	39	Sunki	<i>C. sunki</i>
	VII-27-153	40	Shiikuwasha	<i>C. depressa</i>
	VII-27-154	41	Koji	<i>C. leiocarpa</i>
<i>Pseudofortunella</i>	VIII-28-159	42	Shikikitsu	<i>C. madurensis</i>
<i>Fortunella-Eufortunella</i>				
	102-302	43	Meiwa kumquat	<i>F. crassifolia</i>
	102-304	44	Nagami kumquat	<i>F. margarita</i>
<i>Poncirus</i>				
	103-401	45	Trifoliate orange	<i>P. trifoliata</i>

^aThe classification and nomenclature of Citrus plants are based on Tanaka's system.^b—, not investigated.

Table 2. Flavonoid Concentrations in Citrus Fruits

Category number	Ref.	Name	Flavanones							Flavones										
			ERC	NER	NRT	NRG	HSP	NHP	NPO	PON	RTN	IRF	RFN	DSM	NDM	SNT	NOB	TNG	HPM	
I	1	Cabuyao	0.8	3.3	0.0	0.0	129	285	43.9	492	0.0	0.0	0.0	20.9	57.5	63.0	0.0	4.1	0.0	
II	2	Mexican lime	3.9	0.0	4.4	0.0	197	1.5	3.0	3.3	0.0	10.2	0.0	14.7	3.9	0.0	0.4	0.7	0.0	
	3	Tahiti lime	54.7	0.0	16.1	5.0	351	0.0	2.7	9.7	9.3	17.7	0.0	15.1	5.3	0.6	0.2	0.3	0.0	
	4	Bergamot	10.1	288	14.3	438	2.1	590	2.5	1240	34.8	0.0	43.3	11.9	33.1	0.5	0.2	0.2	0.0	
	5	Biroro	4.7	0.0	10.6	6.0	869	0.0	2.9	1.1	0.0	9.9	0.0	31.0	11.3	1.3	7.1	2.0	1.0	
III	6	Citron	4.5	0.0	0.0	0.0	5.3	0.0	0.0	0.0	3.6	0.0	0.0	4.2	0.0	0.0	0.1	0.1	0.1	
	7	Eureka lemon	102	0.0	11.4	0.0	356	0.0	0.0	0.0	0.0	15.8	1.3	20.8	0.0	0.3	0.0	0.0	0.0	
	8	Sweet lemon	0.2	0.0	16.8	1.9	258	0.0	4.5	0.0	3.2	14.6	0.0	7.0	0.0	0.0	0.0	0.0	0.0	
	9	Lumie	71.8	0.0	0.7	0.0	192	0.0	0.0	0.0	12.0	0.0	0.0	14.8	0.0	0.5	0.0	0.0	1.1	
IV	10	Hirado buntan	0.0	0.0	0.3	617	0.0	7.3	2.6	0.1	1.7	0.0	19.1	0.2	0.9	0.3	0.0	0.6	0.0	
	11	Shaten yu	0.0	0.7	0.0	297	0.0	3.4	0.0	0.9	56.8	0.0	3.2	0.2	2.5	0.1	0.0	0.2	0.0	
	12	Marsh grapefruit	18.3	0.0	170	1360	0.0	21.0	5.3	304	5.1	0.0	9.5	0.0	18.5	0.0	1.0	0.0	0.2	
	13	Kinukawa	5.7	19.3	29.0	173	8.5	177	0.9	24.8	10.2	1.9	16.0	0.0	0.2	0.2	0.7	1.5	0.1	
	14	Hassaku	7.7	4.4	85.2	303	31.4	166	1.0	14.2	3.2	4.0	10.5	0.0	0.9	0.7	0.5	0.9	0.4	
V	15	Natsudaikai	0.0	7.9	26.2	444	11.7	144	0.5	18.9	0.0	0.0	18.3	0.2	0.8	0.3	0.4	1.4	0.0	
	16	Sanboku	38.1	0.0	503	21.8	369	0.0	2.9	1.3	0.0	5.2	0.0	2.8	7.7	0.0	1.7	3.3	1.6	
	17	Sour orange	4.9	210	17.0	979	0.0	684	1.6	282	29.0	2.0	56.6	1.6	17.3	0.0	4.0	2.5	0.0	
	18	Valencia	15.9	2.7	166	0.0	962	0.0	57.1	0.0	10.8	0.3	1.5	1.4	7.7	8.8	5.0	2.2	0.5	
	19	Morita navel	9.6	0.0	178	1.4	642	0.0	34.8	0.0	5.9	0.8	0.7	1.4	10.4	3.2	2.0	1.1	0.7	
	20	Iyo	3.9	0.0	192	0.0	463	0.0	8.8	0.0	0.0	0.4	0.0	0.8	1.3	0.9	4.9	2.3	0.4	
	21	Hyuganatsu	6.5	0.0	135	3.3	186	0.0	36.9	0.0	0.0	0.8	1.6	0.4	4.8	0.5	5.4	6.4	1.8	
	22	Shunkokan	34.8	0.2	1050	0.2	761	0.0	9.6	0.0	1.4	67.0	0.0	9.1	3.1	0.6	8.1	3.6	0.2	
	VI	23	Yuzu	0.5	3.5	129	75.0	122	63.9	2.9	14.4	0.0	1.9	0.3	0.6	7.2	0.6	0.0	0.0	0.0
		24	Sudachi	41.5	16.6	74.0	50.9	31.8	80.1	3.4	1.4	52.3	0.0	1.2	2.5	10.5	0.0	0.0	0.0	0.0
25		Kabosu	0.7	1.2	94.4	37.0	105	36.9	1.6	1.7	55.7	0.0	0.4	0.0	9.2	0.3	0.6	1.5	0.4	
VII	26	King	9.7	0.0	392	1.1	568	0.0	17.2	0.0	9.2	0.3	0.8	0.7	4.2	0.1	2.4	2.3	3.0	
	27	Satsuma	1.4	0.0	134	0.0	412	0.0	15.1	0.0	11.4	1.0	1.1	0.6	0.8	0.0	1.5	1.1	0.6	
	28	Yatsushiro	0.9	0.3	3.6	0.0	306	0.0	1.5	0.0	0.0	0.0	1.4	0.0	0.4	0.6	5.4	5.3	3.1	
	29	Keraji	2.2	0.0	7.5	0.0	242	0.0	2.5	1.2	0.0	0.0	5.3	1.8	5.5	1.8	27.7	35.3	2.2	
	30	Oto	0.0	0.0	3.4	0.0	259	0.0	1.4	0.0	0.0	0.0	0.0	0.0	0.0	0.5	4.4	4.1	2.5	
	31	Ponkan	1.9	0.9	63.4	0.0	594	0.0	21.2	0.0	1.8	3.0	0.0	4.7	1.7	1.8	30.6	33.7	0.0	
	32	Dancy tangerine	0.4	0.9	29.8	0.0	626	0.0	16.9	0.0	0.0	5.5	0.0	3.6	0.0	2.2	33.0	20.9	0.0	
	33	Clementine	1.6	0.0	28.2	0.0	546	0.0	4.0	0.0	3.6	5.5	0.0	8.8	1.0	0.9	2.0	2.1	1.9	
	34	Jimikan	1.3	0.2	21.6	0.0	665	0.0	14.6	0.0	0.0	0.7	0.0	5.0	1.0	1.0	16.5	14.0	0.0	
	35	Shikaikan	0.7	0.0	82.0	0.0	641	0.0	27.0	0.0	2.1	2.7	0.0	3.6	1.1	3.7	44.2	24.0	0.6	
	36	Tachibana	9.1	0.0	6.1	0.0	328	0.0	2.6	0.0	0.0	0.0	0.0	3.3	9.9	6.5	54.3	42.0	0.0	
	37	Kobenimikan	1.4	0.0	10.9	0.0	408	0.0	8.0	2.4	0.0	0.6	0.0	6.6	2.3	0.8	26.9	24.1	0.0	
	38	Kishu	0.0	0.0	63.3	0.0	702	0.0	16.0	0.0	0.0	14.2	1.6	14.6	13.3	1.1	23.5	21.7	0.0	
	39	Sunki	0.0	6.0	57.5	0.0	721	0.0	21.9	3.2	16.7	2.6	0.0	5.9	13.0	24.4	14.8	10.5	0.0	
	40	Shiikuwasha	2.0	0.0	6.0	0.0	411	0.0	3.2	0.0	0.0	0.0	0.5	5.1	0.0	1.8	30.3	18.0	0.0	
41	Koji	84.2	0.0	198	0.0	148	0.0	28.8	0.0	0.0	0.0	0.0	1.4	0.4	1.0	12.2	12.5	0.0		
VIII	42	Shikikitsu	1.8	1.9	313	30.6	48.1	82.5	1.8	0.0	5.3	0.0	2.4	6.3	1.4	0.9	4.3	2.6	0.0	
	102	43	Oval kumquat	0.0	0.0	399	3.1	1.6	0.0	19.5	27.8	0.0	0.0	3.9	0.0	0.0	0.0	0.0	0.0	
	44	Meiwa kumquat	0.0	0.0	279	3.4	0.9	0.0	11.8	23.2	0.0	0.0	4.6	0.0	0.0	0.0	0.0	0.0		
103	45	Trifoliolate orange	0.0	0.0	27.0	379	0.0	0.0	43.5	2410	8.6	2.4	0.0	0.9	36.0	0.4	0.0	0.3	0.0	

Each value is the average of four replications (mg/100 g fresh weight).

Table 3. Flavonoid Concentrations in Peel Tissue of Citrus Fruits

Category number	Ref.	Name	Flavanones							Flavones										
			ERC	NER	NRT	NRG	HSP	NHP	NPO	PON	RTN	IRF	RFN	DSM	NDM	SNT	NOB	TNG	HPM	
I	1	Cabuyao	1.1	4.3	0.0	0.0	64.6	253	21.6	284	0.0	0.0	0.0	19.2	17.5	123	0.0	5.9	0.0	
II	2	Mexican lime	5.6	0.0	6.5	0.0	258	4.9	6.1	13.5	0.0	25.9	0.0	30.7	15.3	1.3	1.7	2.3	0.6	
	3	Tahiti lime	105	0.0	56.3	10.3	1210	0.0	7.7	36.7	6.0	55.3	0.0	52.5	20.4	2.2	1.6	1.0	0.0	
	4	Bergamot	6.6	331	11.1	456	1.2	450	2.0	458	37.2	0.0	65.9	40.0	10.7	1.8	0.6	0.6	0.0	
	5	Biroro	10.3	0.0	10.1	8.0	1310	0.0	6.0	2.5	0.0	21.3	0.0	61.5	14.0	2.9	10.6	4.4	2.2	
III	6	Citron	2.9	0.0	0.0	0.0	1.4	0.0	0.0	0.0	0.6	0.0	0.0	2.6	0.0	0.0	0.1	0.1	0.1	
	7	Eureka lemon	132	0.0	22.5	0.0	711	0.0	0.0	0.0	0.0	35.5	2.9	43.2	0.0	0.7	0.0	0.0	0.0	
	8	Sweet lemon	0.0	0.0	38.8	7.9	491	0.0	12.6	0.0	3.2	49.1	0.0	15.4	0.0	0.0	0.0	0.5	0.0	
	9	Lumie	146	0.0	1.9	0.0	468	0.0	0.0	0.0	27.2	0.0	0.0	47.5	0.0	1.9	0.0	0.0	4.1	
IV	10	Hirado buntan	0.0	4.4	1.0	976	0.0	9.0	4.7	0.0	4.3	0.0	14.0	0.7	3.5	0.0	0.0	0.0	0.0	
	11	Shaten yu	0.0	0.9	0.0	333	0.0	4.5	0.0	2.0	16.6	0.0	2.0	0.0	0.6	0.3	0.0	0.4	0.0	
	12	Marsh grapefruit	9.2	0.0	190	2100	0.0	20.3	8.4	462	5.1	0.0	18.4	0.0	11.0	0.0	4.6	0.0	0.0	
	13	Kinukawa	6.8	34.5	19.3	286	19.8	390	2.5	52.6	9.2	0.0	30.8	0.0	0.7	0.6	2.1	4.3	0.2	
	14	Hassaku	0.3	6.9	70.1	414	35.4	259	1.1	14.9	2.5	0.0	13.9	0.0	2.0	0.0	1.3	2.1	0.9	
V	15	Natsudaidai	0.0	4.7	20.8	639	8.2	271	1.8	39.3	0.0	0.0	16.9	0.7	4.4	1.1	1.5	0.4	0.0	
	16	Sanbokan	65.2	0.0	969	54.9	881	0.0	4.5	3.6	0.0	11.3	0.0	7.6	8.5	0.0	4.6	9.0	4.2	
	17	Sour orange	3.8	220	22.0	1470	0.0	1090	2.7	567	41.3	3.7	108	3.8	43.8	0.0	8.9	5.9	0.0	
	18	Valencia	5.9	0.0	66.5	0.0	1410	0.0	42.1	0.0	0.0	1.1	5.8	5.5	3.0	34.0	18.1	8.5	2.0	
	19	Morita navel	8.1	0.0	228	7.0	2070	0.0	78.0	0.0	0.0	4.2	3.7	7.1	7.8	16.1	10.3	5.5	3.6	
	20	Iyo	7.1	0.0	269	0.0	965	0.0	15.6	0.0	0.0	1.3	0.0	2.6	4.4	2.9	16.5	7.8	1.3	
	21	Hyuganatsu	4.5	0.0	184	0.0	347	0.0	53.7	0.0	0.0	2.1	4.3	1.0	10.6	1.4	13.9	17.2	5.0	
	22	Shunkokan	43.0	0.0	1160	0.0	1270	0.0	8.6	0.0	0.0	9.6	0.0	18.8	8.3	1.3	18.5	8.3	0.5	
	VI	23	Yuzu	0.9	6.9	147	100	192	103	3.3	24.9	0.0	4.3	0.8	1.3	3.0	1.3	0.0	0.2	0.0
		24	Sudachi	65.0	24.9	97.6	70.7	38.9	145	0.3	3.3	121	0.0	2.9	4.5	4.4	0.0	0.0	0.4	0.0
25		Kabosu	1.5	3.3	146	59.6	210	75.3	2.7	4.7	143	0.0	1.1	0.0	5.3	0.8	1.7	4.2	1.1	
VII	26	King	12.5	0.0	478	3.2	1230	0.0	28.1	0.0	2.0	0.7	2.4	2.0	5.6	0.3	6.9	6.7	8.6	
	27	Satsuma	3.3	0.0	281	0.0	1540	0.0	51.5	0.0	4.9	4.4	4.6	2.6	3.3	0.0	6.4	4.8	2.4	
	28	Yatsushiro	0.5	1.4	6.0	0.0	820	0.0	2.6	0.0	0.0	0.0	7.0	0.0	1.9	3.1	22.8	25.9	15.2	
	29	Keraji	3.3	0.0	5.4	0.0	421	0.0	2.4	0.0	0.0	0.0	11.6	4.0	16.7	3.9	53.8	71.3	4.2	
	30	Oto	0.0	0.0	5.9	0.0	627	0.0	1.9	0.0	0.0	0.0	0.0	0.0	0.0	1.8	16.3	15.4	9.4	
	31	Ponkan	4.0	0.5	44.2	0.0	1370	0.0	28.4	0.0	0.0	5.1	0.0	17.6	6.8	6.6	110	124	0.0	
	32	Dancy tangerine	1.6	0.0	70.1	0.0	3970	0.0	57.5	0.0	0.0	7.3	0.0	30.8	0.0	19.1	273	174	0.0	
	33	Clementine	5.4	0.0	57.8	0.0	1800	0.0	10.4	0.0	3.8	19.0	0.0	35.4	4.4	4.0	8.4	8.8	8.2	
	34	Jimikan	3.4	1.2	7.6	0.0	2160	0.0	17.6	0.0	0.0	3.8	0.0	24.5	5.5	5.1	83.5	73.7	0.0	
	35	Shikaikan	0.6	0.0	135	0.0	2150	0.0	85.3	0.0	9.8	12.2	0.0	16.6	4.0	17.2	199	110	2.7	
	36	Tachibana	25.6	0.0	9.8	0.0	749	0.0	3.5	0.0	0.0	0.0	0.0	11.3	3.9	22.1	182	141	0.0	
	37	Kobenimikan	2.1	0.0	14.7	0.0	1190	0.0	12.8	8.9	0.0	2.3	0.0	23.2	11.6	0.8	90.8	84.5	0.0	
	38	Kishu	1.2	0.0	104	0.0	2120	0.0	31.9	0.0	0.0	43.1	5.8	51.5	5.8	4.2	82.9	78.3	0.0	
	39	Sunki	0.5	6.1	56.4	0.0	1480	0.0	23.8	12.2	40.5	8.3	0.0	20.3	13.2	89.1	53.7	38.1	0.0	
	40	Shiikuwasha	4.1	0.0	16.1	0.0	1040	0.0	7.6	0.0	0.0	1.3	1.4	14.4	0.0	4.7	85.2	51.1	0.0	
41	Koji	180	0.0	421	0.0	607	0.0	115	0.0	0.0	0.0	0.0	10.9	2.7	8.0	87.9	94.6	0.0		
VIII	42	Shikikitsu	1.6	6.8	297	16.8	56.3	81.9	5.0	0.0	3.1	0.0	3.3	13.4	4.7	3.3	13.3	7.7	0.0	
102	43	Oval kumquat	0.0	0.0	322	2.3	1.7	0.0	15.5	20.3	0.0	0.0	3.1	0.0	0.0	0.0	0.0	0.0	0.0	
44	Meiwa kumquat	0.0	0.0	199	3.3	1.0	0.0	11.4	13.3	0.0	0.0	0.0	8.0	0.0	0.0	0.0	0.0	0.0		
103	45	Trifoliolate orange	0.0	0.0	49.6	690	0.0	0.0	3.5	1130	17.9	7.8	0.0	2.9	14.0	1.2	0.0	1.1	0.0	

Each value is the average of four replications (mg/100 g fresh weight).

Table 4. Flavonoid Concentrations in Juice Vesicle Tissue of Citrus Fruits

Category number	Ref.	Name	Flavanones							Flavones									
			ERC	NER	NRT	NRG	HSP	NHP	NPO	PON	RTN	IRF	RFN	DSM	NDM	SNT	NOB	TNG	HPM
I	1	Cabuyao	1.1	2.2	0.0	0.0	253	248	74.0	433	0.0	0.0	0.0	31.9	52.3	1.6	0.0	4.5	0.0
II	2	Mexican lime	2.0	0.0	1.4	0.0	42.0	0.0	2.2	0.0	0.0	7.7	0.0	2.2	0.0	0.0	0.0	0.0	0.0
	3	Tahiti lime	47.1	0.0	2.2	4.0	56.0	0.0	1.1	0.0	13.4	5.5	0.0	2.2	0.0	0.0	0.0	0.0	0.0
III	4	Bergamot	13.4	293	17.7	523	2.6	762	3.3	1870	28.3	0.0	39.9	0.0	48.1	0.0	0.0	0.0	0.0
	5	Biroro	0.0	0.0	2.8	5.4	0.0	0.0	0.7	0.0	0.0	1.3	0.0	0.0	8.3	0.0	0.8	0.0	0.0
	6	Citron	10.7	0.0	0.0	0.0	16.0	0.0	0.0	0.0	13.9	0.0	0.0	7.6	0.0	0.0	0.0	0.0	0.0
IV	7	Eureka lemon	81.5	0.0	0.8	0.0	63.8	0.0	0.0	0.0	0.0	1.6	0.0	6.1	0.0	0.0	0.0	0.0	0.0
	8	Sweet lemon	0.0	0.0	4.0	0.0	59.3	0.0	0.0	0.0	4.3	0.0	0.0	2.2	0.0	0.0	0.0	0.0	0.0
	9	Lumie	37.9	0.0	0.0	0.0	57.3	0.0	0.0	0.0	7.7	0.0	0.0	1.2	0.0	0.0	0.0	0.0	0.0
V	10	Hirado buntan	0.0	0.0	0.0	78.5	1.7	1.5	0.0	0.0	0.0	0.0	5.6	0.6	9.4	0.0	0.0	0.0	0.0
	11	Shaten yu	0.0	0.0	0.0	184	0.0	2.6	0.0	0.0	111	0.0	5.7	0.0	5.6	0.0	0.0	0.0	0.0
	12	Marsh grapefruit	27.5	0.0	208	1270	0.0	45.7	4.1	256	5.4	0.0	10.6	0.0	14.6	0.0	0.0	0.0	0.0
	13	Kinukawa	4.4	9.0	34.5	86.1	2.2	50.9	0.0	6.8	12.7	3.5	7.7	0.0	0.0	0.0	0.0	0.0	0.0
	14	Hassaku	15.7	1.6	108	133	29.0	61.1	0.7	5.7	4.7	8.1	8.3	0.0	0.0	0.0	0.0	0.0	0.0
VI	15	Natsudaikai	0.0	1.8	33.6	331	3.1	47.7	0.0	11.2	0.0	0.0	22.3	0.0	0.0	0.0	0.0	0.0	0.0
	16	Sanboku	28.9	0.0	150	0.0	66.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	17	Sour orange	7.2	292	16.5	693	0.0	483	1.1	0.0	32.4	0.0	28.9	0.0	11.9	0.0	0.9	0.0	0.0
	18	Valencia	19.7	5.5	54.3	0.0	93.2	0.0	2.9	0.0	21.6	0.0	0.0	0.0	0.0	0.0	0.6	0.0	0.0
	19	Morita navel	9.9	0.0	120	0.0	94.4	0.0	5.6	0.0	9.3	0.0	0.0	0.0	6.1	0.0	0.0	0.0	0.0
	20	Iyo	2.1	0.0	125	0.0	178	0.0	3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	21	Hyuganatsu	10.6	0.0	97.8	6.1	62.9	0.0	20.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.0	0.0
	22	Shunkokan	6.6	0.0	633	0.0	221	0.0	4.1	0.0	3.8	51.2	0.0	2.3	3.6	0.0	0.0	0.0	0.0
	23	Yuzu	0.8	1.6	330	139	174	93.0	6.8	21.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	24	Sudachi	39.9	17.2	99.2	61.2	55.3	52.7	8.4	0.0	0.0	0.0	0.0	2.3	8.4	0.0	0.0	0.0	0.0
VII	25	Kabosu	0.0	0.0	33.0	10.8	27.4	9.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	26	King	9.2	0.0	311	0.0	153	0.0	7.4	0.0	17.5	0.0	0.0	0.0	2.4	0.0	0.0	0.0	0.0
	27	Satsuma	0.2	0.0	15.4	0.0	8.7	0.0	0.0	0.0	0.0	0.0	0.0	1.4	0.0	0.0	0.0	0.0	0.0
	28	Yatsushiro	0.5	0.0	1.1	0.0	12.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.6	0.0	0.0
	29	Keraji	1.5	0.0	6.2	0.0	31.1	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	6.6	4.8	0.6
	30	Oto	0.0	0.0	0.0	0.0	20.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	31	Ponkan	0.8	0.7	58.8	0.0	84.7	0.0	9.5	0.0	3.2	0.0	0.0	0.0	0.0	0.0	0.8	0.0	0.0
	32	Dancy tangerine	0.0	0.8	7.0	0.0	24.2	0.0	0.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.9	0.0	0.0
	33	Clementine	0.0	0.0	7.4	0.0	37.3	0.0	0.0	0.0	4.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	34	Jimikan	0.0	0.0	13.6	0.0	26.4	0.0	1.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.2	0.0	0.0
	35	Shikaikan	0.7	0.0	25.4	0.0	52.9	0.0	1.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0
	36	Tachibana	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	33.7	0.0	0.0	0.0	0.0
	37	Kobenimikan	1.2	0.0	2.5	0.0	24.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.1	3.2	1.4	0.0
38	Kishu	0.5	0.0	10.0	0.0	20.1	0.0	1.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.8	0.7	0.0	
39	Sunki	1.2	14.1	43.8	0.0	213	0.0	8.0	0.0	17.6	0.0	1.2	0.0	4.3	3.3	2.7	1.7	0.0	
40	Shiikuwasha	1.5	0.0	0.0	0.0	19.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.5	0.8	0.0	
41	Koji	80.4	0.0	167	0.0	74.0	0.0	13.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.8	0.0	0.0	
VIII	42	Shikikitsu	2.8	0.0	402	41.2	53.5	106	0.7	3.9	8.9	0.0	2.9	3.9	0.0	0.0	0.8	0.9	0.0
102	43	Oval kumquat	0.0	0.0	616	0.0	2.4	0.0	26.6	39.9	0.0	0.0	6.7	0.0	0.0	0.0	0.0	0.0	0.0
44	Meiwa kumquat	0.0	0.0	418	3.2	1.7	0.0	24.4	36.8	0.0	0.0	2.5	0.0	0.0	0.0	0.0	0.0	0.0	
103	45	Trifoliolate orange	0.0	0.0	13.0	93.7	0.0	0.0	45.2	1340	7.9	0.0	0.0	0.0	35.5	0.0	0.0	0.0	0.0

Each value is the average of four replications (mg/100 g fresh weight).

Table 5. Flavonoid Concentrations in Flavedo Tissue of Citrus Fruits

Category number	Ref.	Name	Flavanones							Flavones										
			ERC	NER	NRT	NRG	HSP	NHP	NPO	PON	RTN	IRF	RFN	DSM	NDM	SNT	NOB	TNG	HPM	
I	1	Cabuyao	4.5	4.2	0.0	0.0	30.2	75.8	8.3	42.3	0.0	0.0	0.0	34.3	21.8	396	nd	6.1	0.0	
II	2	Mexican lime	8.2	0.0	5.9	0.0	125	0.0	8.4	25.3	0.0	18.8	0.0	24.9	24.6	2.4	1.9	1.8	1.1	
	3	Tahiti lime	122	0.0	17.6	13.7	462	0.0	2.3	71.8	13.3	13.3	0.0	43.5	40.5	4.8	1.8	2.1	0.0	
	4	Bergamot	6.8	183	7.2	155	0.0	115	1.8	6.8	89.7	0.0	129	82.5	6.3	4.3	0.7	1.3	0.0	
	5	Biroro	12.7	0.0	4.4	11.5	660	0.0	2.6	6.3	0.0	12.4	0.0	57.2	14.9	4.7	23.0	9.2	5.6	
III	6	Citron	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	4.6	0.0	0.0	1.6	0.0	0.0	1.0	0.9	1.1	
	7	Eureka lemon	69.4	0.0	2.7	0.0	193	0.0	0.0	0.0	0.0	10.0	5.8	37.7	0.0	2.5	0.0	0.0	0.0	
	8	Sweet lemon	0.0	0.0	13.7	6.6	228	0.0	5.7	0.0	6.2	35.0	0.0	14.9	0.0	0.0	0.0	0.5	0.0	
	9	Lumie	81.0	0.0	0.0	0.0	70.8	0.0	0.0	0.0	56.8	0.0	0.0	19.0	0.0	3.9	0.0	0.0	8.7	
IV	10	Hirado buntan	0.0	0.0	2.6	136	0.0	7.4	1.5	1.9	5.3	0.0	10.5	3.6	3.0	2.1	0.0	7.8	0.0	
	11	Shaten yu	0.0	0.0	0.0	15.9	0.0	0.0	0.0	10.7	0.0	0.0	2.0	0.0	3.0	1.6	0.0	1.9	0.0	
	12	Marsh grapefruit	25.8	0.0	114	994	0.0	29.5	5.1	140	0.0	0.0	46.9	0.0	6.3	0.0	11.1	0.0	2.2	
	13	Kinukawa	9.5	28.7	6.4	61.4	3.1	157	2.3	52.4	14.1	0.0	50.0	0.0	2.5	2.2	5.4	12.6	0.8	
	14	Hassaku	1.1	1.8	15.8	45.9	16.1	84.0	0.9	6.0	9.3	0.0	5.7	0.0	7.6	0.0	4.8	7.8	3.3	
V	15	Natsudaidai	0.0	0.0	7.7	102	1.1	113	2.2	43.5	0.0	0.0	4.4	2.1	2.9	3.3	2.4	10.2	0.0	
	16	Sanbokan	33.5	0.0	121	3.1	228	0.0	1.7	16.3	0.0	7.3	0.0	10.0	9.8	0.0	13.5	26.4	11.8	
	17	Sour orange	3.4	144	9.3	520	0.0	569	2.0	661	63.7	11.8	25.1	7.3	35.5	0.0	25.3	16.3	0.0	
	18	Valencia	8.2	0.0	13.3	0.0	495	0.0	10.1	0.0	0.0	2.3	11.8	11.1	6.0	64.6	33.7	15.7	4.1	
	19	Morita navel	17.4	0.0	40.0	12.0	1170	0.0	18.2	0.0	0.0	9.7	8.7	16.6	10.5	34.5	22.0	11.4	8.3	
	20	Iyo	10.3	0.0	65.2	0.0	363	0.0	3.6	0.0	0.0	3.6	0.0	7.5	12.9	3.5	38.9	18.7	3.7	
	21	Hyuganatsu	12.2	0.0	36.0	0.0	100	0.0	6.9	0.0	0.0	0.0	15.0	3.4	22.6	4.8	42.0	50.3	14.8	
	22	Shunkokan	57.5	0.0	12.8	0.0	1230	0.0	0.0	0.0	0.0	51.0	0.0	41.0	31.9	12.1	139	64.5	4.5	
	VI	23	Yuzu	1.1	15.1	52.4	50.4	40.7	69.6	1.3	27.6	0.0	3.3	2.0	3.4	8.0	3.4	0.0	0.6	0.0
		24	Sudachi	26.9	21.9	18.8	20.4	5.4	40.8	3.3	0.0	4.7	0.0	4.1	6.6	3.6	0.0	0.0	0.8	0.0
25		Kabosu	0.0	0.0	17.9	15.7	21.6	24.1	3.0	0.0	0.0	0.0	2.6	0.0	8.3	1.9	4.0	8.5	2.8	
VII	26	King	0.0	0.0	34.0	6.9	3500	0.0	4.9	0.0	7.1	2.5	8.3	6.9	2.1	1.1	19.2	17.9	23.0	
	27	Satsuma	2.0	0.0	74.5	0.0	945	0.0	19.5	0.0	12.6	4.8	10.1	4.4	3.5	2.2	10.8	7.6	3.8	
	28	Yatsushiro	0.9	2.4	4.0	0.0	355	0.0	1.1	0.0	0.0	0.0	10.8	0.0	3.3	4.2	36.8	41.1	24.4	
	29	Keraji	8.7	0.0	5.8	0.0	89.4	0.0	0.0	0.0	0.0	0.0	9.4	7.4	14.3	7.0	98.7	134	7.2	
	30	Oto	0.0	0.0	4.4	0.0	237	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.8	23.6	22.0	13.4	
	31	Ponkan	5.1	0.9	15.0	0.0	812	0.0	5.3	0.0	0.0	6.1	0.0	29.4	7.5	11.6	198	218	0.0	
	32	Dancy tangerine	0.0	0.0	38.8	0.0	2680	0.0	27.3	0.0	0.0	12.1	0.0	53.4	0.0	35.0	507	315	0.0	
	33	Clementine	1.7	0.0	4.8	0.0	760	0.0	1.5	0.0	7.5	7.8	0.0	37.2	4.7	5.9	14.0	14.8	13.7	
	34	Jimikan	6.9	2.5	1.8	0.0	1750	0.0	8.3	0.0	0.0	7.8	0.0	42.7	11.2	10.5	160	138	0.0	
	35	Shikaikan	0.0	0.0	37.5	0.0	1590	0.0	20.3	0.0	16.7	20.9	0.0	26.1	3.5	27.1	315	172	4.7	
	36	Tachibana	30.2	0.0	8.0	0.0	549	0.0	2.2	0.0	0.0	0.0	0.0	13.6	5.4	28.0	233	179	0.0	
	37	Kobenimikan	3.8	0.0	3.7	0.0	413	0.0	1.6	8.0	0.0	2.0	0.0	3.8	9.0	2.2	166	149	0.0	
	38	Kishu	0.0	0.0	42.9	0.0	1800	0.0	13.7	0.0	0.0	34.0	8.8	64.4	3.3	5.9	118	108	0.0	
	39	Sunki	0.0	0.0	30.8	0.0	1220	0.0	9.0	13.2	37.9	9.4	0.0	27.4	6.9	121	67.4	48.8	0.0	
	40	Shiikuwasha	0.0	0.0	4.6	0.0	616	0.0	0.9	0.0	0.0	2.4	2.5	21.3	0.0	6.4	122	71.3	0.0	
41	Koji	156	0.0	273	0.0	263	0.0	32.3	0.0	0.0	0.0	0.0	14.4	4.5	11.6	128	136	0.0		
VIII	42	Shikikitsu	1.3	9.9	254	0.0	33.3	55.6	5.7	0.0	6.0	0.0	5.0	15.2	5.7	6.4	23.0	13.3	0.0	
	102	43	Oval kumquat	0.0	0.0	340	5.3	0.0	0.0	17.7	16.3	0.0	0.0	4.7	0.0	0.0	0.0	0.0	0.0	
	44	Meiwa kumquat	0.0	0.0	205	6.3	0.0	0.0	12.2	9.0	0.0	0.0	1.5	0.0	0.0	0.0	0.0	0.0		
103	45	Trifoliolate orange	0.0	0.0	27.5	192	0.0	0.0	5.1	48.7	35.3	11.4	0.0	3.0	4.2	2.4	0.0	2.1	0.0	

Each value is the average of four replications (mg/100 g fresh weight).

Table 6. Flavonoid Concentrations in Albedo Tissue of Citrus Fruits

Category number	Ref.	Name	Flavanones							Flavones										
			ERC	NER	NRT	NRG	HSP	NHP	NPO	PON	RTN	IRF	RFN	DSM	NDM	SNT	NOB	TNG	HPM	
I	1	Cabuyao	0.0	4.3	0.0	0.0	76.2	313	26.1	365	0.0	0.0	0.0	14.1	16.1	30.8	0.0	5.8	0.0	
II	2	Mexican lime	2.7	0.0	7.2	0.0	409	10.5	3.4	0.0	0.0	34.0	0.0	37.3	4.8	0.0	1.5	2.8	0.0	
	3	Tahiti lime	90.8	0.0	88.3	7.4	1840	0.0	12.2	7.7	0.0	90.1	0.0	60.0	3.7	0.0	1.3	0.0	0.0	
	4	Bergamot	6.3	435	13.9	670	2.1	686	2.2	777	0.0	0.0	21.0	9.9	13.8	0.0	0.5	0.0	0.0	
	5	Biroro	8.8	0.0	13.7	5.7	1730	0.0	8.3	0.0	0.0	27.0	0.0	64.3	13.5	1.7	2.6	1.3	0.0	
III	6	Citron	3.3	0.0	0.0	0.0	1.6	0.0	0.0	0.0	0.0	0.0	0.0	2.8	0.0	0.0	0.0	0.0	0.0	
	7	Eureka lemon	156	0.0	30.4	0.0	918	0.0	0.0	0.0	0.0	45.7	1.7	45.4	0.0	0.0	0.0	0.0	0.0	
	8	Sweet lemon	0.0	0.0	66.4	9.3	780	0.0	20.1	0.0	0.0	64.5	0.0	16.1	0.0	0.0	0.0	0.6	0.0	
	9	Lumie	206	0.0	3.7	0.0	833	0.0	0.0	0.0	0.0	0.0	0.0	73.7	0.0	0.0	0.0	0.0	0.0	
IV	10	Hirado buntan	0.0	0.0	1.5	1400	1.1	12.0	6.4	0.0	0.0	0.0	16.1	0.0	3.7	0.0	0.0	0.0	0.0	
	11	Shaten yu	0.0	1.2	0.0	407	0.0	5.5	0.0	0.0	20.5	0.0	2.0	0.0	0.0	0.0	0.0	0.0	0.0	
	12	Marsh grapefruit	0.0	0.0	231	2700	0.0	15.2	10.2	638	7.9	0.0	2.7	0.0	13.6	0.0	1.1	0.0	0.0	
	13	Kinukawa	5.9	36.6	23.9	366	25.7	474	2.6	52.6	7.4	0.0	23.9	0.0	0.0	0.0	0.9	1.3	0.0	
	14	Hassaku	0.0	8.8	90.0	549	42.4	324	1.2	18.1	0.0	0.0	16.9	0.0	0.0	0.0	0.0	0.0	0.0	
V	15	Natsudaidai	0.0	7.1	27.7	919	12.0	353	1.5	37.1	0.0	0.0	23.4	0.0	5.2	0.0	1.0	1.4	0.0	
	16	Sanbokan	74.3	0.0	1210	69.8	1070	0.0	5.3	0.0	0.0	12.5	0.0	6.9	8.1	0.0	2.1	4.0	2.1	
	17	Sour orange	3.9	255	27.9	1900	0.0	1320	3.0	524	31.1	0.0	146	2.2	47.6	0.0	1.4	1.1	0.0	
	18	Valencia	3.6	0.0	118	0.0	2300	0.0	73.0	0.0	0.0	0.0	0.0	0.0	0.0	4.5	3.1	1.6	0.0	
	19	Morita navel	1.1	0.0	370	3.2	2740	0.0	123	0.0	0.0	0.0	0.0	0.0	5.7	2.1	1.4	1.0	0.0	
	20	Iyo	5.4	0.0	376	0.0	1280	0.0	21.9	0.0	0.0	0.0	0.0	0.0	0.0	2.1	4.7	2.1	0.0	
	21	Hyuganatsu	1.4	0.0	244	0.0	446	0.0	72.5	0.0	0.0	3.0	0.0	0.0	5.8	0.0	2.6	3.8	1.1	
	22	Shunkokan	41.3	0.0	1290	0.0	1270	0.0	9.6	0.0	0.0	101	0.0	16.2	5.5	0.0	4.2	1.6	0.0	
	VI	23	Yuzu	0.7	1.9	205	130	283	124	4.5	23.2	0.0	4.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0
		24	Sudachi	99.5	27.5	169	116	69.3	240	2.8	6.2	226	0.0	1.8	2.5	5.0	0.0	0.0	0.0	0.0
25		Kabosu	2.7	5.7	240	91.6	348	113	2.5	8.1	247	0.0	0.0	0.0	3.0	0.0	0.0	1.1	0.0	
VII	26	King	17.6	0.0	659	1.7	1580	0.0	37.5	0.0	0.0	0.0	0.0	7.1	0.0	2.0	2.1	2.8		
	27	Satsuma	2.8	0.0	494	0.0	2100	0.0	87.1	0.0	0.0	2.9	2.6	2.0	3.0	0.0	1.8	1.9	1.0	
	28	Yatsushiro	0.0	0.0	8.6	0.0	1430	0.0	4.6	0.0	0.0	0.0	2.0	0.0	1.8	4.4	5.9	3.3		
	29	Keraji	0.0	0.0	5.1	0.0	617	0.0	3.8	0.0	0.0	0.0	12.9	2.0	18.1	2.1	27.2	34.0	2.5	
	30	Oto	0.0	0.0	8.6	0.0	1310	0.0	5.2	0.0	0.0	0.0	0.0	0.0	0.0	3.4	3.7	2.2		
	31	Ponkan	3.1	0.0	69.8	0.0	1870	0.0	48.7	0.0	0.0	4.2	0.0	7.2	6.2	2.2	33.3	41.5	0.0	
	32	Dancy tangerine	2.8	0.0	94.2	0.0	4970	0.0	80.8	0.0	0.0	3.7	0.0	13.4	0.0	7.0	93.1	65.1	0.0	
	33	Clementine	9.2	0.0	112	0.0	2870	0.0	19.6	0.0	0.0	30.5	0.0	33.4	4.1	2.0	2.5	2.6	2.6	
	34	Jimikan	0.0	0.0	13.3	0.0	2560	0.0	26.4	0.0	0.0	0.0	0.0	7.2	0.0	0.0	10.8	12.6	0.0	
	35	Shikaikan	1.4	0.0	274	0.0	2940	0.0	177	0.0	0.0	0.0	0.0	3.1	4.8	3.3	35.9	21.3	0.0	
	36	Tachibana	13.1	0.0	14.5	0.0	1280	0.0	7.0	0.0	0.0	0.0	0.0	5.1	0.0	6.2	46.7	39.9	0.0	
	37	Kobenimikan	1.0	0.0	21.5	0.0	1680	0.0	19.8	9.5	0.0	2.4	0.0	14.4	13.2	0.0	43.7	44.5	0.0	
	38	Kishu	2.7	0.0	186	0.0	2530	0.0	56.1	0.0	0.0	55.1	1.9	34.3	9.0	2.0	37.1	39.6	0.0	
	39	Sunki	1.3	16.6	100	0.0	1940	0.0	49.0	10.7	45.1	6.3	0.0	8.2	24.0	34.5	30.3	19.8	0.0	
	40	Shiikuwasha	9.0	0.0	30.0	0.0	1550	0.0	15.7	0.0	0.0	0.0	0.0	6.1	0.0	2.5	41.0	26.9	0.0	
41	Koji	180	0.0	647	0.0	1160	0.0	242	0.0	0.0	0.0	0.0	5.6	0.0	2.5	26.8	31.0	0.0		
VIII	42	Shikikitsu	1.9	3.5	343	0.0	80.8	110	4.3	0.0	0.0	1.6	11.5	3.7	0.0	2.9	1.7	0.0		
102	43	Oval kumquat	0.0	0.0	307	0.0	3.0	0.0	13.7	23.3	0.0	0.0	1.9	0.0	0.0	0.0	0.0	0.0		
44	Meiwa kumquat	0.0	0.0	192	0.0	2.1	0.0	10.6	18.0	0.0	0.0	14.9	0.0	0.0	0.0	0.0	0.0	0.0		
103	45	Trifoliolate orange	0.0	0.0	72.2	1200	0.0	0.0	1.8	2240	0.0	4.1	0.0	2.9	24.1	0.0	0.0	0.0		

Each value is the average of four replications (mg/100 g fresh weight).

Table 7. Flavonoid Concentrations in Segment Epidermis Tissue of Citrus Fruits

Category number	Ref.	Name	Flavanones							Flavones									
			ERC	NER	NRT	NRG	HSP	NHP	NPO	PON	RTN	IRF	RFN	DSM	NDM	SNT	NOB	TNG	HPM
I	1	Cabuyao	0.0	3.6	0.0	0.0	116	463	56.5	1080	0.0	0.0	0.0	14.4	44.0	45.5	0.0	1.2	0.0
II	2	Mexican lime	7.0	0.0	9.4	0.0	502	1.3	1.6	0.0	0.0	0.0	26.6	0.0	0.0	0.0	0.6	0.0	
	3	Tahiti lime	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	4	Bergamot	3.9	189	7.0	7.0	1.8	156	0.0	247	65.3	0.0	6.4	0.0	4.3	0.0	0.0	0.0	
	5	Biroro	0.0	0.0	18.7	4.0	952	0.0	0.0	0.0	0.0	0.0	11.1	2.8	0.0	7.4	0.0	0.0	
III	6	Citron	0.9	0.0	0.0	0.0	9.3	0.0	0.0	0.0	0.0	0.0	8.7	0.0	0.0	0.0	0.0	0.0	
	7	Eureka lemon	129	0.0	21.2	0.0	479	0.0	0.0	0.0	12.5	1.3	12.7	0.0	0.0	0.0	0.0	0.0	
	8	Sweet lemon	0.0	0.0	26.7	0.0	554	0.0	7.5	0.0	0.0	13.7	0.0	10.9	0.0	0.0	0.0	0.0	
	9	Lumie	110	0.0	2.2	0.0	372	0.0	0.0	0.0	0.0	0.0	15.8	0.0	0.0	0.0	0.0	0.0	
IV	10	Hirado buntan	0.0	0.0	1.4	1120	1.3	6.0	5.2	0.0	0.0	6.1	0.0	0.0	1.4	0.0	0.0	0.0	
	11	Shaten yu	0.0	2.7	0.0	670	0.0	3.7	0.0	0.0	37.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	12	Marsh grapefruit	13.6	0.0	118	1130	0.0	16.2	5.5	301	5.5	0.0	2.1	0.0	5.7	0.0	0.0	0.0	
	13	Kimukawa	11.4	27.9	39.9	310	4.1	150	0.0	32.1	0.0	12.6	0.0	0.0	0.0	0.0	0.0	0.0	
	14	Hassaku	4.1	7.8	65.7	765	34.8	317	2.0	61.5	0.0	2.9	9.0	0.0	0.0	0.0	0.0	0.0	
V	15	Natsudaikai	0.0	60.3	0.0	661	82.4	408	0.0	9.2	0.0	0.0	0.0	0.0	0.0	0.0	1.0	0.0	
	16	Sanboku	3.1	0.0	766	17.4	149	0.0	12.9	0.0	10.7	0.0	0.0	12.9	0.0	0.0	0.0	0.0	
	17	Sour orange	5.7	148	13.7	746	0.0	391	0.8	205	8.6	1.9	9.5	0.0	2.2	0.0	0.0	0.0	
	18	Valencia	9.2	0.0	244	0.0	1100	0.0	89.8	0.0	0.0	0.0	0.0	6.6	0.0	0.0	0.0	0.0	
	19	Morita navel	9.7	0.0	333	0.0	1010	0.0	92.7	0.0	0.0	0.0	0.0	6.2	0.0	0.0	0.0	0.0	
	20	Iyo	5.4	0.0	357	0.0	668	0.0	22.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	21	Hyuganatsu	1.4	0.0	157	4.2	198	0.0	53.6	0.0	0.0	0.0	0.0	1.8	0.0	0.0	0.0	0.0	
	22	Shunkokan	85.7	1.6	1920	1.3	774	0.0	27.7	0.0	0.0	35.1	0.0	0.0	0.0	0.0	0.0	0.0	
	VI	23	Yuzu	0.0	1.1	121	65.3	75.5	33.3	2.9	4.4	0.0	0.0	0.0	3.5	0.0	0.0	0.0	0.0
		24	Sudachi	12.9	6.0	26.5	19.1	3.8	15.1	0.0	0.0	0.0	0.0	0.0	3.5	0.0	0.0	0.0	0.0
25		Kabosu	0.7	0.0	123	48.0	80.8	25.9	2.3	0.0	16.0	0.0	0.0	4.6	0.0	0.0	0.0	0.0	
VII	26	King	5.9	0.0	515	0.0	456	0.0	25.8	0.0	0.0	0.0	0.0	2.7	0.0	0.0	0.0	0.0	
	27	Satsuma	1.8	0.0	290	0.0	423	0.0	34.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	28	Yatsushiro	1.9	0.0	6.9	0.0	466	0.0	3.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	29	Keraji	1.0	0.0	20.5	0.0	288	0.0	8.1	0.0	0.0	0.0	0.0	0.0	0.0	3.5	5.3	0.0	
	30	Oto	0.0	0.0	9.7	0.0	439	0.0	4.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
	31	Ponkan	2.6	2.5	133	0.0	1340	0.0	61.4	0.0	0.0	12.7	0.0	0.0	0.0	3.3	2.7	0.0	
	32	Dancy tangerine	1.0	2.1	97.3	0.0	831	0.0	55.8	0.0	0.0	26.7	0.0	0.0	0.0	3.2	3.4	0.0	
	33	Clementine	2.0	0.0	68.3	0.0	669	0.0	10.3	0.0	0.0	6.8	0.0	3.3	0.0	0.0	0.0	0.0	
	34	Jimikan	3.0	0.0	58.8	0.0	1180	0.0	50.0	0.0	0.0	0.0	1.8	0.0	0.0	0.7	0.7	0.0	
	35	Shikaikan	1.2	0.0	17.1	0.0	627	0.0	32.2	0.0	0.0	0.0	0.0	0.0	0.0	1.9	0.9	0.0	
	36	Tachibana	5.4	0.0	11.4	0.0	380	0.0	5.7	0.0	0.0	0.0	0.0	0.0	0.0	1.9	1.3	0.0	
	37	Kobenimikan	0.9	0.0	45.4	0.0	570	0.0	37.2	0.0	0.0	0.0	1.8	0.0	0.0	3.0	2.8	0.0	
	38	Kishu	0.0	0.0	19.3	0.0	811	0.0	43.0	0.0	0.0	17.3	0.0	5.5	12.3	0.0	2.1	2.2	
39	Sunki	1.5	4.0	121	0.0	1080	0.0	51.8	0.0	7.6	1.9	0.0	2.5	5.4	2.6	1.3	1.2		
40	Shiikuwasha	0.0	0.0	3.6	0.0	337	0.0	4.4	0.0	0.0	0.0	1.3	0.0	1.2	2.1	1.3	0.0		
41	Koji	45.5	0.0	198	0.0	123	0.0	34.8	0.0	0.0	0.0	0.0	0.0	0.0	1.2	1.2	0.0		
VIII	42	Shikikitsu	0.0	0.0	204	38.0	39.2	44.8	0.0	0.0	0.0	0.0	3.7	0.0	0.0	0.6	0.0	0.0	
102	43	Oval kumquat	0.0	0.0	716	8.4	1.5	0.0	37.5	57.8	0.0	6.8	0.0	0.0	0.0	0.0	0.0	0.0	
	44	Meiwa kumquat	0.0	0.0	339	5.3	0.0	0.0	1.2	31.2	0.0	0.0	3.3	0.0	0.0	0.0	0.0	0.0	
103	45	Trifoliolate orange	0.0	0.0	17.8	329	0.0	0.0	62.6	3890	0.0	0.0	0.0	24.9	0.0	0.0	0.0	0.0	

Each value is the average of four replications (mg/100 g fresh weight).

Eureka lemon (#7), species of *Aurantium* (#16, 18–20, 22) and *Acrumen* (VII) sections (Table 2). The albedo contained the highest concentration among these fruits, except for King (#26) in which the concentration of HSP in the flavedo was more than twice that in the albedo (Table 5 and 6). In contrast, the HSP concentration in the juice vesicle was the lowest among the fruit tissues in these species (Table 4). All species tested in the *Cephalocitrus* (IV) and *Osmocitrus* (VI) sections contained NHP, while those in the *Citrophorum* (III) and *Acrumen* sections (VII) didn't (Table 2). The species containing NRG at more than 10 mg/100 g of fruit contained NHP, except for Sanbokan (#16) and Trifoliate orange (#45) (Table 2). Bergamot (#4) and Sour orange (#17) contained NHP at high concentrations in the juice vesicle (Table 4). Valencia (#18), Morita navel (#19), and Hyuganatsu (#21) belonging to the *Aurantium* section (VII), as well as Cabuyao (#1) and Trifoliate orange (#45), contained NPO at a high concentration in fruit of the species tested (Table 2). The concentrations of this compound in the segment epidermis were the same or higher than in the peel of these species (Tables 3 and 7). Trifoliate orange (#45) contained PON at very high concentrations in the segment epidermis, albedo, juice vesicle, and flavedo: 3890, 2240, 1340, and 48.7 mg/100 g, respectively. Cabuyao (#1), species in the *Limonellus* (II), *Cephalocitrus* (IV), part of *Aurantium* (#15–17), and *Osmocitrus* (VI) sections, and those in *Fortunella* (102) also contained this flavonoid. RTN was widely distributed in *Citrus* and *Poncirus* (103) fruits, although no specific trend was apparent, except that all species in the *Cephalocitrus* (IV) section contained it. Shunkokan (#22) contained IRF at exceptionally high concentrations in the albedo, juice vesicle, flavedo, and segment epidermis: 101, 51.2, 51.0, and 35.1 mg/100 g, respectively (Tables 4–7). The species with high concentrations of RFN were Bergamot (#4), Sour orange (#17), Natsudaikai (#15), and those in the *Cephalocitrus* section (IV) (Table 2). The concentration of this compound was lowest in the segment epidermis among the fruit tissues (Table 7). DSM widely occurred in all species except those of the *Cephalocitrus* (IV) section and *Fortunella* (102) species (Table 2). The concentrations of DSM tended to be higher in *Limonellus* (II) and *Citrophorum* (III) fruits than in other fruits. NDM was widely distributed in all tested fruits except for the *Citrophorum* (III) and *Fortunella* (102) species (Table 2). Cabuyao (#1), Bergamot (#4), and Trifoliate orange (#45) in particular contained relatively large amounts of NDM among the fruits tested, especially in the juice vesicle (Table 4). SNT occurred in most fruits tested, except for the *Fortunella* (102) species. Among them Cabuyao (#1) and Sunki (#39) contained it in large quantities in the flavedo (Table 2 and 5). This compound was scarce in the juice vesicle (Table 4) as reported previously.²⁶⁾ NOB was present at high concentrations in species of the *Acrumen* (VII) and *Aurantium* (V) sections (Table 2) but

not in the *Fortunella* (102) and *Poncirus* (103) species. As was the case with SNT, NOB was distributed mostly in the flavedo (Table 5). The distribution pattern of TNG resembled that of NOB, the highest concentration being found in the flavedo of Dancy tangerine (#32), followed by that of Ponkan (#31) (Table 5). HPM occurred in the flavedo of some species in the *Acrumen-Euacrumen* (VII) and *Aurantium* (V) groups at relatively high concentrations, but the levels in the tested species were lower than those of other polymethoxylated flavones (PMFs).

Principal component analysis

The principal component and factor loading values for the flavonoids in each part of the fruits of the 45 species are listed in Table 8. All neohesperidosyl flavanones (NER, NRG, NHP, and PON) investigated and RFN (Fig. 1), a neohesperidosyl flavone, had absolute values of more than 0.5 in the 1st principal component (PC), except for PON in the segment epidermis. Another neohesperidosyl flavone, NDM, also had large absolute values, except in the flavedo and segment epidermis. On the other hand, the rutinosyl flavonoids (ERC, NRT, HSP, NPO, IRF, and DSM) except for RTN had opposite scores to those of the neohesperidosyl flavonoids in the 1st PC of each part of the fruits, except for NPO and DSM in the juice vesicle (Tables 7 and 8). From these results, the sugar type was suggested to largely contribute to the 1st PC. Albach and Redman have shown that *Citrus* species could be differentiated on the basis of their content of neohesperidosides or rutinosides.⁶⁾ Kawai *et al.* have reported a multivariate analysis of 66 *Citrus* species, using composition data for 21 flavonoids in the dried edible part which consisted of the juice vesicle and segment epidermis.²⁷⁾ Our results partly agree with their data in that some of the neohesperidosyl flavonoids had a high factor loading on the 1st PC, although the opposing score between the rutinosides and neohesperidosides was not apparent. Concerning the 2nd PC, the PC values for the flavonoid glycosides generally decreased with decreasing polarity of the fruit (Table 8). In this component, the PMFs had negative values, except for HPM. Therefore, the polarity of the flavonoid constituents is suggested to contribute in the 2nd PC of the fruit. In respect of the PC of the segment epidermis, the flavonoid glycosides had inverse values to those of the fruit, and the contribution of PMFs was smaller than that of the fruit. In the analyses of the peel, flavedo and albedo, NOB and TNG in addition to HSP each had a relatively high factor loading among the compounds tested. On the other hand, values for the flavonoids in the juice vesicle had different patterns from those in the other parts of fruits, and no clear trend was apparent. According to the report by Kawai *et al.*, the results of a PCA and cluster analysis of 23 flavonoid compositions in 34 citrus juices were not entirely in agreement with Tanaka's classification.¹⁶⁾ They presumed the composition data were influenced by flavo-

Table 8. Principal Components and Factor Loadings for the Flavonoids in Fruit Tissues of 45 Species

	Principal component											
	Fruit		Peel		Juice vesicle		Flavedo		Albedo		Segment epidermis	
	1	2	1	2	1	2	1	2	1	2	1	2
Eigenvalue	4.47	2.51	4.70	2.56	4.21	3.22	4.25	2.84	4.33	2.57	3.56	2.63
Proportion %	26.27	14.78	27.62	15.04	24.77	18.96	25.01	16.74	25.50	15.15	22.25	16.41
Cumulative %	26.27	41.05	27.62	42.66	24.77	43.72	25.01	41.75	25.50	40.64	22.25	38.66
Factor loading												
ERC	-0.10	0.20	-0.18	0.02	0.03	0.12	-0.06	0.10	-0.21	-0.34	0.18	-0.18
NER	0.81	-0.06	0.72	-0.40	-0.86	0.17	0.82	0.39	0.68	0.23	-0.65	-0.11
NRT	-0.24	0.21	-0.16	0.23	0.13	0.05	-0.14	-0.16	-0.22	-0.25	0.28	-0.17
NRG	0.71	0.16	0.71	-0.10	-0.64	0.26	0.59	0.04	0.73	0.10	-0.63	-0.11
HSP	-0.63	-0.33	-0.66	-0.55	0.17	-0.62	-0.49	0.57	-0.66	0.46	0.76	0.05
NHP	0.87	-0.15	0.82	-0.35	-0.93	-0.04	0.81	0.24	0.83	0.24	-0.66	0.41
NPO	-0.15	-0.66	-0.47	-0.40	-0.23	-0.73	-0.42	0.40	-0.43	0.40	0.46	0.49
PON	0.53	-0.40	0.65	-0.22	-0.78	-0.14	0.75	0.25	0.55	0.12	-0.19	0.60
RTN	0.52	0.21	0.33	0.06	-0.29	0.20	0.70	0.33	0.16	-0.12	-0.49	-0.15
IRF	-0.25	0.07	-0.22	-0.17	0.12	-0.05	-0.16	0.51	-0.23	-0.36	0.45	-0.08
RFN	0.86	0.11	0.84	-0.38	-0.87	0.26	0.64	0.21	0.77	0.28	-0.68	-0.22
DSM	-0.07	-0.38	-0.24	-0.43	-0.17	-0.77	-0.04	0.73	-0.24	-0.28	0.13	0.28
NDM	0.54	-0.74	0.59	-0.46	-0.74	-0.45	0.34	0.30	0.66	0.42	-0.15	0.93
SNT	0.07	-0.77	-0.10	-0.29	-0.03	-0.58	-0.03	0.12	-0.10	0.43	-0.15	0.84
NOB	-0.43	-0.39	-0.53	-0.71	0.20	-0.33	-0.50	0.71	-0.48	0.79	0.48	0.03
TNG	-0.41	-0.40	-0.53	-0.68	-0.01	-0.85	-0.51	0.65	-0.47	0.77	0.42	0.19
HPM	-0.26	0.17	-0.18	0.28	0.12	-0.32	-0.15	-0.24	-0.14	-0.11	—	—

—, not analyzed.

noid localization, pulp content, and water content. Mouly *et al.* have demonstrated the differentiation of sweet oranges, grapefruits, and lemons purchased at a local market by using the pattern recognition technique of 6 the flavanone glycoside composition of juice.¹⁵⁾ Provided that the number of *Citrus* biotypes is limited, the flavonoid composition should enable their differentiation.

The scatter diagrams from the principal component analysis based on the concentration of flavonoids in the fruit samples are shown in Figs. 2(A) and (B). The spots for Cabuyao (#1), Bergamot (#4), Marsh grapefruit (#12), Sour orange (#17), Shunkokan (#22), and Trifoliolate orange (#45) are widely distributed away from the other groups (Fig. 2(A)). The spots for Sour orange (#17) and Bergamot (#4) are relatively close together, agreeing with the report in which Bergamot (#4) was indicated to be a hybrid of Citron (#6) and Sour orange (#17) by using RAPD and SCAR markers.¹⁸⁾ In the *Limonellus* (II) section, the spots for Biroro (#5) and for Bergamot (#4) separated from those of Mexican lime (#2) and Tahiti lime (#3) (Fig. 2(B)). In the *Citrophorum* section (III), the spots for Sweet lemon (#8) and Lumie (#9) are close to each other, but that for Citron (#6) is close to the *Fortunella* (102) area and that for Eureka lemon (#7) is close to the *Acrumen* (VII) area. Excepting the Marsh grapefruit (#12), the species in the *Cephaecitrus* (IV) section formed a group with Natsuda-dai (#15), Sudachi (#24), and Kabosu (#25). The spots for the *Aurantium* (V) species are widely dispersed; that for Natsuda-dai (#15) is located in the *Cephaecitrus* group (IV), those for Sanbokan (#16) and

Iyo (#20) are in the *Acrumen* group (VII-1), those for Valencia (#18), Morita navel (#19), and Hyuganatsu (#21) are relatively near to the *Acrumen* group (VII-1-3), and those for Sour orange (#17) and Shunkokan (#22) are away from those of the other species. The species in the *Aurantium* (V) section are considered to be hybrids of *C. reticulata* and *C. grandis*.²⁸⁾ This difference in flavonoid composition being considered to involve the predominance of the genotype of their parentage. As already mentioned, two spots for the *Osmocitrus* (VI) species appeared within the group of the *Cephaecitrus* (IV) species, and the spot for Yuzu (#23) was away from these. Swingle has defined Yuzu (#23) as a hybrid of *C. ichangensis* and Sunki (#39), and Sudachi (#24) as a hybrid of *C. ichangensis* and *C. grandis*.²⁵⁾ According to the report by Berhow *et al.*, Yuzu (#23) had NRG in the leaves which contained more varieties of phenolic compounds than the fruit, while *C. ichangensis* did not.²⁹⁾ As Sunki (#39) is a mandarin-type fruit which does not contain NRG (Table 2), Yuzu (#23) might have been derived from an additional crossing with other species containing NRG. Species in the *Acrumen* (VII) section commonly include rutynosyl flavonoids such as NRT, HSP and NPO, but not neohesperidosyl ones (Table 2). The 16 species investigated in this section are split into 3 groups, in addition to the spots for Satsuma (#27) and Sunki (#39) which are located apart from them (Fig. 2-(B)). It is apparent that the species belonging to the VII-1 group contained lower levels of both NOB and TNG than those in the VII-2 or VIII-3 group (Table 2). Spots for the *Euacrumen* subsection species (#26-28)

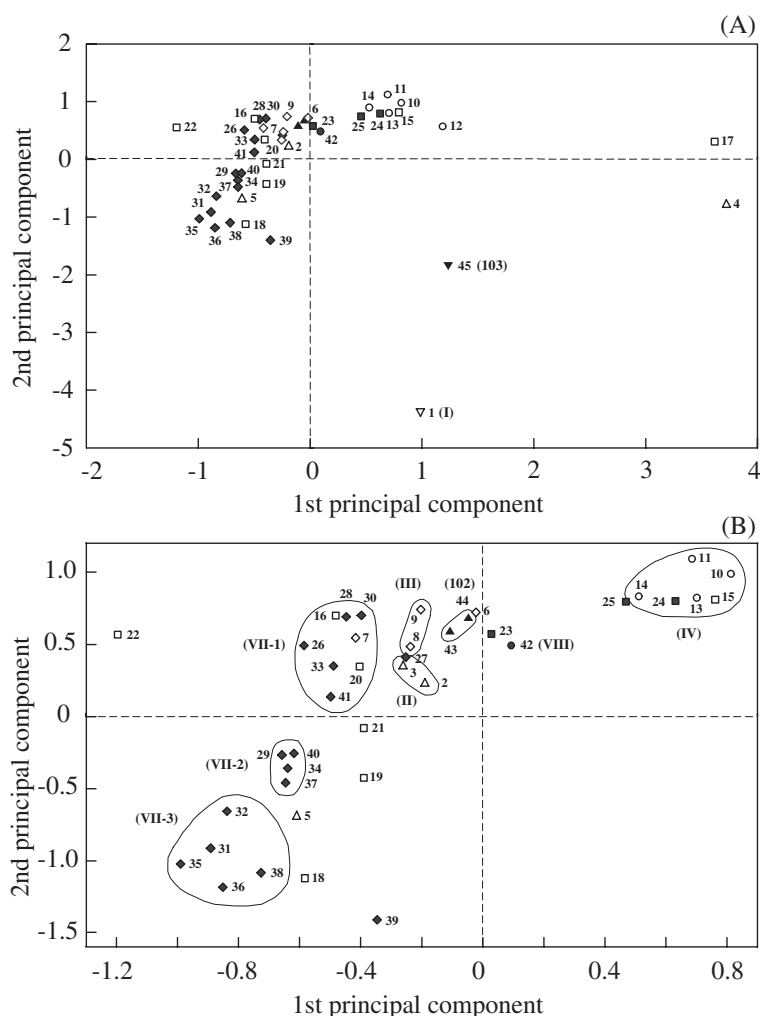


Fig. 2. Scatter Diagram from a Principal Component Analysis Based on the Concentration of Flavonoids in Fruit (A) and a Close-up Detail of the Plot (B).

Symbols: ∇ , *Papeda* (I); Δ , *Limonellus* (II); \diamond , *Citrophorum* (III); \circ , *Cephacitrus* (IV); \square , *Aurantium* (V); \blacksquare , *Osmocitrus* (VI); \blacklozenge , *Acrumen* (VII); \bullet , *Pseudofortunella* (VIII); \blacktriangle , *Fortunella* (102); \blacktriangledown , *Poncirus* (103). Roman numerals and numbers in parentheses indicate Tanaka's category number. See Table 1 for the identification of numbers.

are relatively close to each other near the VII-1 area. The three split groups of the *Acrumen* (VII) species did not seem to have relevance to Tanaka's further classification of this section. Gaydou *et al.* have examined the differentiation of oranges (*C. sinensis*) and mandarins (*C. reticulata*) by using the 6 PMF composition data of fruit peel.¹⁴ In their report, cultivars of *C. sinensis* in *Aurantium* (V) section and *C. reticulata* in *Acrumen* (VII) section were apparently differentiated from each other, although Clementine (#33) and Dancy tangerine (#32), both of which are *Acrumen* (VII) species, were found in the *Aurantium* (V) group. In our experiment, two cultivars of *C. sinensis* (#18 and #19) were separated from the *Acrumen* group, but Sanbokan (#16) and Iyo (#20), both *Aurantium* species, were located in the VII-1 area (Fig. 2).

The dendrogram calculated by the UPGMA method based on the standardized data for flavonoid concentration is shown in Fig. 3. Sour orange (#17), Bergamot

(#4), Cabuyao (#1), Shunkokan (#22), Trifoliolate orange (#45), Marsh grapefruit (#12), and Biroro (#5) are separated from the other species. The species in the *Cephacitrus* (IV) section and Natsudaidai (#15), which include NRG and NHP, are associated with each other, except for Marsh grapefruit (#12) and Syaten yu (#11). In the *Aurantium* (V) section, Valencia (#18), Morita navel (#19) and Hyuganatsu (#21) are associated with each other to a slight degree, but Sambokan (#16) and Iyo (#20), as well as Sour orange (#17), Shunkokan (#22) and Natsudaidai (#15) are separated from these species. Concerning the *Osmocitrus* (VI) species, Yuzu (#23) is separated from Sudachi (#24) and Kabosu (#25), while Yuzu (#23) is associated with Citron (#6), *Fortunella* (102) species and shikikitsu (#42), as is the case with the scatter diagram (Fig. 2(B)). The spots for Sudachi (#24) and Kabosu (#25) are in the *Cephacitrus* (IV) area of the scatter diagram (Fig. 2(B)), although these species are separated from *Cephacitrus* (IV)

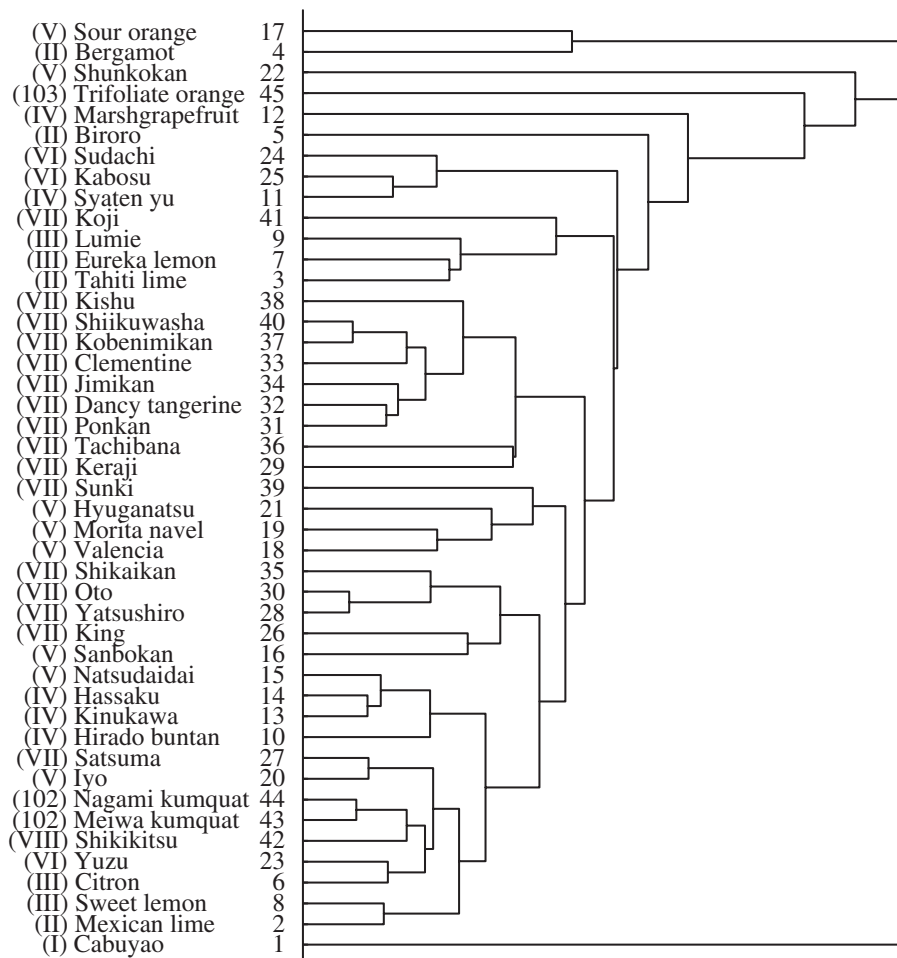


Fig. 3. Dendrogram Calculated by the UPGMA Method Based on the Concentration of Flavonoids in *Citrus* Fruits. Roman numerals and numbers in parentheses indicate Tanaka's category number. See Table 1 for the identification of numbers.

species except Syaten yu (#11). The species in the *Acrumen* (VII) section are roughly separated into two groups, one of them consisting of the *Microacrumen* group species and the other of the *Euacrumen* and *Microacrumen* species. Further classification of these groups did not coincide with the degree of association. Species in the genus *Fortunella* (102) and Shikikitsu (#42), belonging to the *Pseudofortunella* (VIII) section, were associated with each other.

In conclusion, the flavonoid composition in the tissues of 45 citrus species has been revealed for the effective utilization of citrus flavonoids. The flavonoid composition of citrus fruits classified by Tanaka's system was in approximate agreement within each section, except in the *Aurantium* (V) section and those with a peculiar flavonoid composition. The classification of *Citrus* species may differ with the recognition of morphological differences. The development of research in molecular marker techniques would solve this problem.

Acknowledgments

We thank Dr. T. Yoshida and Dr. Y. Ikoma (National Institute of Fruit Tree Science) for providing fruit

samples and Dr. M. Berhow (US Department of Agriculture, Agricultural Research Service) for presenting neodiosmin, as well as for helpful discussions regarding the analytical treatment.

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