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Esters and Glucosides of Hydroxycinnamic Acids in Vegetables

Michael Winter and Karl Herrmann*

Quinic, tartaric, and malic acid esters as well as glucose esters and the glucosides of hydroxycinnamic acids have been determined qualitatively and quantitatively by HPLC in tomatoes (two states of ripeness (green and red) and different types), bell pepper, eggplant, spinach, mangold, beetroot, pea, bush bean, broad bean, lettuce (outer and inner leaves separated), endive, and chicory.

INTRODUCTION

Generally naturally occurring derivatives of hydroxycinnamic acids are represented by their quinic acid or glucose esters, which often have been detected in fruit (Herrmann, 1978; Möller and Herrmann, 1983; Reschke and Herrmann, 1981; Schuster and Herrmann, 1985) and vegetables (Herrmann, 1978; Reschke and Herrmann, 1982; Brandl and Herrmann, 1983a, 1983b).

Additionally the malic acid esters of these phenolic acids could be detected in vegetables, although these compounds were found rarely in nature. Phaseolic acid (caffeoylmalic acid) represents the most detected compound of these derivatives. It has been found in beans (Scarpati and Oriente, 1960) and *Trifolium pratense* (Yoshihara et al., 1974). All of the four hydroxycinnamoyl malic acids have been isolated from small radish (Brandl et al., 1984).

Furthermore, there is another kind of hydroxycinnamic acid ester derived from tartaric acid. These compounds were found frequently in vegetables and fruits. The analysis of these derivatives will be more difficult because of the different naturally occurring stereoisomers like L- and meso-tartaric acid. L-Tartaric acid esters were found in grapes (Ong and Nagel, 1978), wine (Okamura and Watanabe, 1981), cider (Whiting and Coggins, 1975), and chicory (Scarpati and d'Amico, 1960).

The meso-tartaric acid ester of *p*-coumaric acid was found in spinach (Suzuki et al., 1970; Tadera et al., 1970; Oettmeier and Heupel, 1972a, 1972b).

Additionally, dihydroxycinnamoyltartaric acids like dicaffeoyltartaric acid were detected in lettuce (Feucht et al., 1971), endive (Wöldecke and Herrmann, 1974), and chicory (Scarpati and Oriente, 1958).

The glucosides of hydroxycinnamic acids are less distributed. In vegetables they only were found in tomatoes (Fleuriet and Macheix, 1980, 1981; Winter and Herrmann, 1984).

To get more informed about the occurrence of these hydroxycinnamic acid derivatives, especially the glucose derivatives, we have analyzed several important species of vegetables quantitatively and qualitatively by means of HPLC.

Besides the flavonoids, which meanwhile are used as proofs for adulteration in food analysis (Siewek et al., 1984, 1985), the derivatives of hydroxycinnamic acids represent another group of interesting phenolic compounds in plants. Within the biochemical anabolism in plants these substances are precursors of the flavonoids.

Therefore, it will be imaginable that in the future they as well will be used as coindicators for proof of adulteration (Herrmann, 1979). Most of these compounds are of a sufficient stability so that they can be detected even in products like wine and must (Okamura and Watanabe, 1981; Singleton et al., 1978).

The use of hydroxycinnamic acid derivatives as proofs for adulteration will assume their sufficient determination in fruits and vegetables and there in several varieties.

MATERIAL AND METHODS

Standards. The IUPAC nomenclature is used for the quinates (e.g., chlorogenic acid (former 3-*O*-caffeoylquinic acid) is the 5' ester and neochlorogenic acid (the former 5-*O*-caffeoylquinic acid) now will be the 3' ester).

Chlorogenic acid is the only commercially available substance of the hydroxycinnamic acid derivatives (Roth, Karlsruhe, G.F.R.). 5'-Feruloylquinic acid was isolated from green coffee beans and 3'-*p*-coumaroylquinic acid was received from unripe Morello cherries by preparative HPLC. The isomerization of these compounds was carried out by the method of Scarpati and Esposito (1964).

Sinapoylglucose was received by Brandl and Herrmann (1983a) by preparative HPLC from garden cress. Feruloylglucose was isolated by Reschke and Herrmann (1982) from chive, and *p*-coumaroylglucose was placed at our disposal by Birkofer. Caffeoylglucose was isolated by Koeppen and Herrmann (1977). The glucosides of caffeic acid, *p*-coumaric acid, and ferulic acid were synthesized

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at our laboratory by the methods of Mauthner (1912, 1918), Hann (1934), and Helferich and Vorsatz (1936).

Caffeoyltartaric acid was placed at our disposal by Nagel. Wöldecke and Herrmann (1974) isolated dicaffeoyltartaric acid from endive. *p*-Coumaroyl-*meso*-tartaric acid was isolated from spinach leaves by a similar method used by Suzuki et al. (1970).

The malic acid esters of caffeic, *p*-coumaric, ferulic, and sinapic acid were isolated by Brandl et al. (1984).

Analytical HPLC of Hydroxycinnamic Acid Derivatives. The chromatographic analyses were performed with an HPLC system consisting of a pump (type LC-XPD), a gradient programmer (type LC-XPD), a UV detector (type LC-UV), or a diode array detector (Hewlett-Packard, Waldbronn, G.F.R.) and an autosampler (type LCX) (all LC types from Philips, Pye Unicam, Kassel, F.R.G.). The injector valve (Rheodyne, type 7125; Berkeley, CA) was combined with a 10- μ L sample loop. Quantitative results were obtained with a reporting integrator 3390A (Hewlett-Packard, Waldbronn, G.F.R.)

The analyses were made with a 250 \times 4.6 mm i.d. stainless-steel column of Ultrasphere ODS, 5 μ m. Gradient elution was carried out with solvent A (10% methanol in 2% acetic acid) and solvent B (methanol), from 0% B in A to 29% B in A within 50 min at a flow rate of 1.0 mL/min. UV detection was set at 320 nm with a band width of 8 nm.

Preparation of Sample. All vegetables investigated were harvested fully ripe (exceptions are marked in the tables) and in a comparable state of maturity. After harvesting, the vegetables were kept deep-frozen until use. Sample preparation was done by homogenization with a sufficient amount of 80% methanol. The sample was extracted twice while stirring at room temperature for 30 min. The combined extracts were evaporated under reduced pressure to an aqueous residue (100 mL; original extract).

Polyamide Preparation. An aliquot of this original extract was purified and pre-separated by column chromatography on polyamide (250 \times 27 mm i.d.) (sample cleanup). The elution sequence was 150 mL of water, 500 mL of methanol, and 300 mL of methanol containing 2% formic acid. The methanolic fraction led to the glucose derivatives of the hydroxycinnamic acids; with MeOH/formic acid the quinates, tartrates, and malates were obtained. Quantitative analyses of this polyamide pre-separation method were made by Winter and Herrmann (1984, 1986).

Dihydroxycinnamoyltartrates could not be eluted from the column with these eluents. For the determination of these compounds we used the original extract without pre-separation on polyamide.

RESULTS AND DISCUSSION

The quantitative determination of the hydroxycinnamic acid esters was carried out by using the calibration graphs of the corresponding free acids. This method of calibration was necessary, because only a few amounts of these derivatives were available for comparison. On the other hand, Möller and Herrmann (1982) described that the UV responses of the esters and free acids showed only small differences. Brandl and Herrmann (1983c) showed that the determination of chlorogenic acid using the calibration graph of caffeic acid will cause only a negligible error of calculation.

As the determination of the glucosides of the hydroxycinnamic acids took place 30 nm besides their real maximum of absorption, we controlled and confirmed the linearity of their calibration graphs at 320 nm.

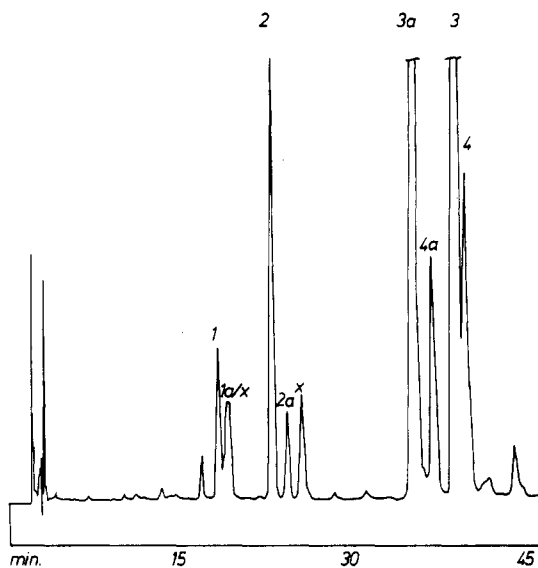


Figure 1. Spinach (methanol/formic acid extract of the polyamide column): 1, *p*-coumaroylglucose; 2, feruloylglucose; 3, *p*-coumaroyl-*meso*-tartaric acid; 4, *p*-coumaroylmalic acid; 5, caffeoylmalic acid; 6, feruloylmalic acid; 7, sinapoylmalic acid; 8, monocaffeoyltartaric acid; 9, 5'-caffeoylquinic acid; 10, dicaffeoyltartaric acid.

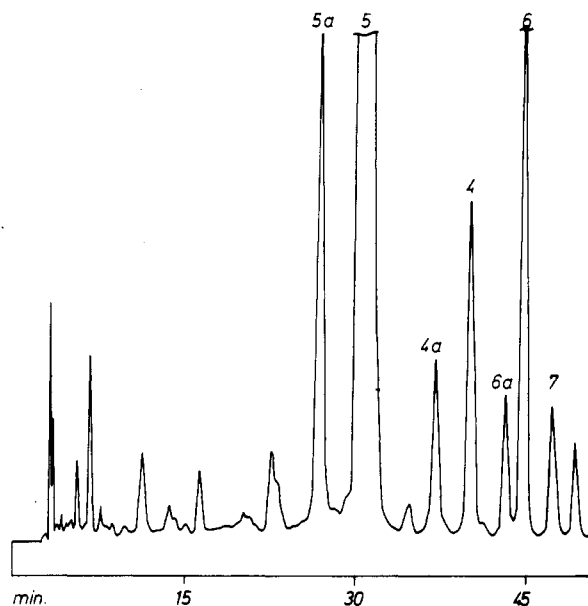


Figure 2. Leaves of bush beans (methanol/formic acid extract of the polyamide column). For peak assignments, see Figure 1.

The qualitative determination was carried out by three different methods. The first criterion was the retention time. The second one was made by cochromatography (the addition of the standard to the extract caused an enlargement of the area of the unknown peak at a positive identification). The third method used for the identification was the possibility of taking the UV spectra of the eluting peak without stopping the flow. This was only possible with a diode array detector.

Our investigations on tomatoes included nine varieties of different types of tomatoes. Additionally we took into consideration the changes of concentrations of the compounds investigated during two steps of ripeness (green and red). Usually chlorogenic acid was detected in higher amounts in the unripe green fruits than in ripe ones. On the other hand, the contents of glucosides increased as well as the 4'- and 3'-caffeoylquinates. With exception of *p*-coumaroylglucose (Table I) the contents of the glucose

Table I. Contents (mg/kg, Fresh Weight) of Hydroxycinnamoyl Derivatives in Tomatoes^a

variety	state or ripeness	5'-CafQ	4'-CafQ	3'-CafQ	CafG	pCoupG	FerG
Pulp Tomato							
Baroso	green	46	+	-	+	+	+
	red	31	6	+	+	+	+
Dombo	green	27	+	-	+	+	+
	red	12	+	-	+	+	+
Yellow Pulpous							
Goldene Königin	green	52	+	-	-	-	-
	red	17	5	-	+	19	+
VSB 3212	green	64	+	-	+	+	+
	red	71	10	+	+	7	+
Normal Type							
Ostona	green	38	+	-	+	+	+
	red	25	6	-	+	7	+
Bellina	green	57	-	-	+	+	+
	red	26	6	-	+	6	+
Ronald	red	23	7	-	+	+	+
Hellfrucht	red	22	7	-	+	+	+
Haubner's Vollendung	red	20	11	-	+	+	+

variety	state of ripeness	Cafglu	pCoupglu	Ferglu
Pulp Tomato				
Baroso	green	6	+	+
	red	25	28	8
Dombo	green	5	+	+
	red	15	25	8
Yellow Pulpous				
Goldene Königin	green	6	+	+
	red	30	62	11
VSB 3212	green	17	10	+
	red	48	68	13
Normal Type				
Ostona	green	16	+	+
	red	39	41	12
Bellina	green	13	10	+
	red	37	42	15
Ronald	red	29	19	10
Hellfrucht	red	21	20	9
Haubner's Vollendung	red	29	28	12

^a Assignment: + = trace, content lower than 2 ppm; - = not detectable; CafQ = caffeoylquinic acid; CafG = 1-*O*-caffeoyl- β -D-glucose; pCoupG = 1-*O*-*p*-coumaroyl- β -D-glucose; FerG = 1-*O*-feruloyl- β -D-glucose; Cafglu = caffeic acid 4-*O*- β -D-glucoside; pCoupglu = *p*-coumaric acid *O*- β -D-glucoside; Ferglu = ferulic acid *O*- β -D-glucoside.

Table II. Contents (mg/kg, Fresh Weight) of Hydroxycinnamoyl Derivatives in Bell Pepper and Eggplant^a

variety	state of ripeness	5'-CafQ	4'-CafQ	3'-CafQ	5'-pCoupQ	5'-FerQ
Bell Pepper						
Bruinsma wonder	green	-	-	-	-	-
	red	-	-	-	-	-
Goldstar	green	-	-	-	-	-
	yellow	-	-	-	-	-
Eggplant						
from Italy	ripe	632	11	+	+	17
from Spain	ripe	575	8	+	+	15

variety	state of ripeness	CafG	pCoupG	FerG	SinG	Cafglu	pCoupglu	Ferglu
Bell Pepper								
Bruinsma wonder	green	+	-	+	-	-	-	-
	red	+	-	11	5	-	-	-
Goldstar	green	+	-	+	-	-	-	-
	yellow	+	-	+	+	-	-	-
Eggplant								
from Italy	ripe	+	-	+	+	-	-	-
from Spain	ripe	+	-	+	+	-	-	-

^a Assignment: + = trace, content lower than 2 ppm; - = not detectable; pCoupQ = *p*-Coumaroylquinic acid; FerQ = feruloylquinic acid; SinG = 1-*O*-sinapoyl- β -D-glucose; additional assignments, see Table I.

esters seemed to be constant.

The other vegetables belonging to the Solanaceae like bell pepper or eggplant (Table II) showed no similarity in

their profile of compounds compared with tomatoes.

All of the Chenopodiaceae investigated—spinach, man- gold, and beetroot—contained feruloylglucose. The most

Table III. Contents (mg/kg, Fesh Weight) of Hydroxycinnamoyl Derivatives of Some Chenopodiaceae Vegetables^a

variety	3'-CafQ	3'-pCoumQ	3'-FerQ	pCoumG	FerG	SinG	pCoumM	pCoumM
Spinach								
Nores	-	-	-	18	42	-	189	29
Jaguar	-	-	-	21	42	-	223	23
Symphony	-	-	-	16	64	-	230	25
Mangold								
Rex (stalk)	-	-	-	-	18	-	-	-
Rex (leaves)	-	-	-	3	88	14	-	-
Lukullus (stalk)	-	-	-	-	14	-	-	-
Lukullus (leaves)	-	-	-	-	52	6	-	-
Beetroot								
Dardani (leaves)	2	+	-	+	44	8	-	-
Dardani (stalk)	5	+	2	-	18	+	-	-
Dardani (tuber)	5	3	5	-	+	-	-	-
Ruval (leaves)	4	+	-	+	30	7	-	-
Ruval (stalk)	+	+	-	-	12	+	-	-
Ruval (tuber)	+	+	-	+	+	-	-	-

^a Assignment: pCoumM = *p*-Coumaroyl-*meso*-tartaric acid; pCoumM = *p*-coumaroylmalic acid; additional assignments, see Tables I and II. Brandl (unpublished) found in the leaves (tuber) of beetroot "Rote Kugel": 3'-CafQ, 5 (1); 3'-pCoumQ, 2 (-); 3'-FerQ, 2 (-); FerG, 40 (3); SinG, 5 (-).

Table IV. Contents (mg/kg, Fresh Weight) of Hydroxycinnamoyl Derivatives of Some Fabaceae Vegetables^a

variety	CafM	pCoum	FerM	SinM	4'-pCoumQ	CafG	pCoumG	FerG	SinG
Pea									
Wunder v. Kelvedon (leaves)	-	-	-	-	-	5	7	11	4
Wunder v. Kelvedon (hull)	-	-	-	-	-	-	3	-	-
Wunder v. Kelvedon (seed)	-	-	-	-	4	-	-	-	-
Noblesse (hull)	-	-	-	-	-	-	7	15	5
Noblesse (seed)	-	-	-	-	72	-	-	10	-
Bush Bean									
Fanion (leaves)	939	32	53	13	-	-	-	-	-
Broad Bean									
Hedosa (leaves)	87	166	10	-	-	-	-	17	5
Hedosa (hull)	9	61	31	-	-	-	-	2	3
Hedosa (seed)	-	+	-	-	-	-	-	-	-
Osna grün (leaves)	162	164	10	-	-	-	-	18	13
Osna grün (hull)	+	+	+	-	-	-	-	+	4
Osna grün (seed)	-	+	-	-	-	-	-	-	-

variety	Cafglu	pCoumglu	Ferglu
Pea			
Wunder v. Kelvedon (leaves)	9	-	-
Wunder v. Kelvedon (hull)	2	-	-
Wunder v. Kelvedon (seed)	-	-	-
Noblesse (hull)	5	-	-
Noblesse (seed)	-	-	-
Bush Bean			
Fanion (leaves)	-	-	-
Broad Bean			
Hedosa (leaves)	3	+	5
Hedosa (hull)	+	+	3
Hedosa (seed)	-	-	-
Osna green (leaves)	3	+	+
Osna green (hull)	+	+	2
Osna green (seed)	-	-	-

^a Assignment: CafM = caffeoylmalic acid; FerM = feruloylmalic acid; SinM = sinapoylmalic acid; additional assignments, see Tables I-III.

interesting compound was *p*-coumaroyl-*meso*-tartaric acid detected in spinach leaves in large amounts (180-230 ppm) (Figure 1). Mangold (stalks, leaves) only showed glucose esters of *p*-coumaric, caffeic, and ferulic acid. The main compounds of beetroot also have been the glucose esters besides the 3' isomers of *p*-coumaroyl-, caffeoyl-, and feruloylquinates (Table III).

The proportion of hydroxycinnamic acid derivatives in Fabaceae is shown in Table IV. None of these tested derivatives could be found in the seed of bush bean. Only a trace of *p*-coumaroylmalate was detected in broad bean.

More different hydroxycinnamate compounds of these Fabaceae could be detected in their leaves or hulls (Figure 2).

Table V shows the results of the investigations of Chi-oriaceae. The whole pattern of caffeoyl derivatives with the exception of the glucose derivatives was found in lettuce (Figure 3). The different investigations on lettuce showed that there were no difference between greenhouse and outdoor cultivation. Relative to the detected compounds we found a gradient of falling concentration from the outer to the inner leaves of lettuce.

Table V. Contents (mg/kg, Fresh Weight) of Hydroxycinnamoyl Derivatives in Some Chicoriaceae Vegetables^a

variety		CafT	DiCafT	CafM	FerM	5'-CafQ	FerG
Lettuce							
Salina	A (a)	27	154	32	-	64	-
Salina	I (a)	15	96	3	-	11	-
Sitonia	A (a)	28	167	21	-	45	-
Sitonia	I (a)	20	105	4	-	18	-
Corelli	A (b)	10	38	15	-	22	-
Corelli	I (b)	17	74	2	-	8	-
Capitan	A (b)	12	62	18	-	39	-
Capitan	I (b)	11	43	+	-	5	-
Endive							
Jeti		21	163	10	14	36	-
Diva		31	334	13	16	124	-
Chicory							
from Belgium		4	17	-	-	5	+

^aAssignment: A = outer leaves of lettuce (green); I = inner leaves of lettuce (yellow green); (a) = greenhouse cultivation; (b) = outdoor cultivation; CafT = caffeoyltartaric acid; DiCafT = di-caffeoyltartaric acid; additional assignments, see Tables I-IV.

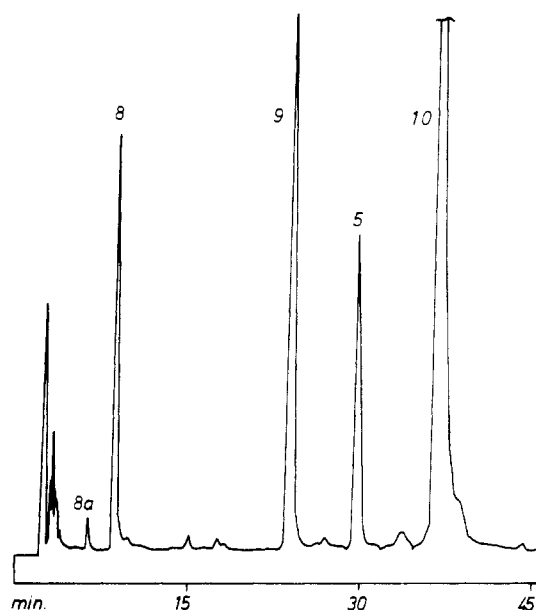


Figure 3. Lettuce (original extract). For peak assignments, see Figure 1.

Our investigations show that each of the species has its own pattern of compounds. Within a species of vegetables with different varieties, the profile remained constant while amounts vary only.

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Registry No. 5'-CafQ, 327-97-9; 4'-CafQ, 35906-35-5; 3'-CafQ, 906-33-2; CafG, 14364-08-0; Cafglu, 17093-82-2; CafM, 39015-77-5;

CafT, 1234-09-9; diCafT, 6537-80-0; pCouglu, 14364-05-7; 5'-pCougQ, 32451-86-8; pCougM, 39015-75-3; 3'-pCougQ, 1899-30-5; 4'-pCougQ, 53539-37-0; pCougG, 7139-64-2; pCougMT, 37551-71-6; FerG, 7196-71-6; Ferglu, 14364-12-6; 5'-FerQ, 40242-06-6; 3'-FerQ, 53905-80-9; FerM, 39015-76-4; SinM, 76656-81-0; sinapoylglucose, 14364-09-1.

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