Effects of Protein Supplements on Carbonyl Compounds and Flavor in Bread

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ABSTRACT

The contents of volatile carbonyl compounds were determined in legume and oilseed flours and protein concentrates, and in protein-supplemented doughs and breads. 2-Propanone, 2-butanone, 3-methylbutanal, and hexanal were tentatively identified as the principal carbonyl compounds in soy flour and sunflower, fababean, and field pea protein concentrates while large quantities of ethanal also appeared in the fermented doughs. Bread crumbs and crusts contained high levels of the same five carbonyl compounds plus furfural and hydroxymethylfurfural, generally in higher concentrations than the all-wheat control. The flavor ratings for the protein-supplemented breads decreased with increasing contents of total carbonyl compounds and ranked the products in the general order of wheat > sunflower > soy = fababean > field pea.

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Considerable attention has been focused on the use of legume and oilseed flours and protein concentrates to improve the nutritional value of wheat bread (Cotton 1974, D'Appolonia 1977, Fleming and Sosulski 1977, Hallab et al 1974, Matthews et al 1970). While some commercial success has been achieved, acceptance of nonwheat protein supplements has been limited because of undesirable effects on dough and baking properties, and organoleptic characteristics. Adverse flavors are a major limitation in the use of soy (Cotton 1974), fababean (Patel et al 1977), field pea, and sunflower (Sosulski and Fleming 1979) proteins in baked products.

Volatile and nonvolatile compounds in bread that serve as flavor stimuli include acids, alcohols, aldehydes, esters, ether derivatives, furan derivatives, hydrocarbons, ketones, lactone derivatives, pyrazines, pyrrole derivatives, and sulfur compounds (Coffman 1967, Maga 1974). Volatile carbonyl compounds have been identified as the principal aroma and flavor compounds in wheat and rye breads (Linko et al 1962, Lorenz and Maga 1972a, Ng et al 1960, Rao et al 1978). However, the carbonyl compounds in legume and oilseed protein-supplemented breads, which exhibit distinct flavors (Sosulski and Fleming 1979), have not been investigated.

The objective of the present study was to determine the quantitative composition of the major volatile carbonyl compounds in protein supplements, fermented doughs, crumbs, and crusts of protein-supplemented breads. Products investigated included soy flour, sunflower, fababean, and field pea protein concentrates. Sensory evaluations of the aroma and taste of the baked products also were done.

MATERIALS AND METHODS

Wheat Flour and Protein Supplements

The wheat flour used was an untreated baker's straight grade (72% extraction) flour; the vital gluten was supplied by Industrial Grain Products Ltd., Montreal. The wheat flour and vital gluten were extracted from commercial hard red spring wheat and contained 15.4 and 89.4%, respectively, of crude protein (N×6.25) and 0.6 and 0.9% respectively, of ash, dry basis. Regular defatted and "Bland 50" soy flours were obtained from Staley Manufacturing Co., Decatur, IL. The fababean (*Vicia faba minor*) and field pea (*Pisum sativum*) protein concentrates were prepared by pin milling the dehulled seeds and air classifying to separate the starch and protein fractions (Vose et al 1976). Sunflower concentrate (*Helianthus annuus*) was prepared by diffusing cracked dehulled kernels in an aqueous medium (pH 4.5) for 4 hr at 60°C to remove chlorogenic acid (Sosulski et al 1973). The dried seed meal was extracted with Skellysolve F (petroleum ether, b.p. 40–60°C), desolventized, and ground to pass through a 100-mesh Tyler sieve. The fababean, field pea, and sunflower concentrates contained 61.1, 56.5, and 76.0%, respectively, of crude protein, whereas the soy flour had 50.5%.

Bread-Making Procedure

The composite flour formulations included ratios of wheat flour/protein supplement/vital gluten of 83:15:2 for soy, fababean, and field pea bread, and 86:12:2 for sunflower bread to provide breads with approximately 22.5% protein, dry basis. The bread-making formula included 100.0 g wheat flour or composite flour (14% moisture), 4.0 g nonfat dry milk, 3.0 g compressed yeast, 5.0 g sucrose, 1.75 g salt, 0.19 g ammonium phosphate monobasic, 10 ppm potassium bromate, 0.3 g malt syrup, and water as needed. Where composite flours were used, 1.0 g polyoxyethylene-8-stearate/100 g composite flour, as dough conditioner, was added to the flour ingredients prior to mixing the dough. The dough was mixed by the straight-dough AACC procedure and baked for 25 min at 220°C in an experimental rotary baking oven. Fermented dough samples were taken for analysis after proofing for 180 min.

Carbonyl Determination

For determination of carbonyl compounds, 50 g portions (dry basis) of wheat flour, protein supplement, ground freeze-dried dough, bread crumb, and crust were extracted with successive 100, 75, and 50 ml portions of carbonyl-free chloroform in a Sorvall Omnimixer. The combined chloroform extract was added to 500 ml of 1.0% 2,4-dinitrophenylhydrazine (DNPH) reagent in sulfuric acid and the mixture refluxed at 50°C for 2 hr to form hydrazones (El Dash and Johnson 1970). The chloroform layer containing the hydrazones was removed using a separatory funnel and the DNPH reagent layer was extracted twice with 25-ml aliquots of carbonyl-free chloroform. The combined chloroform extract of hydrazones was mixed with 1.0 g anhydrous sodium sulfate to absorb water and then concentrated under vacuum. The concentrated extract was filtered to remove sodium sulfate, and adjusted to 25 ml volume with chloroform.

A flash exchange chromatography procedure (Stephen and Teszler 1960), as modified by Jones and Monroe (1965), was adopted for the regeneration of the carbonyl compounds from their DNPH derivatives and for their subsequent separation and determination. Twenty-five milligrams of Celite was mixed with 0.5 to 1.0 ml of chloroform containing the hydrazones and the mixture was dried in an air oven at 60°C. Thirty-five milligrams of a mixture of *p*-dimethylaminobenzaldehyde and oxalic acid (DMAB/oxalic acid, 6.0:5.3 w/w) as exchange reagent (Jones and Monroe 1965) was mixed with the dry Celite and hydrazones. An accurately weighed sample of approximately 10 mg of this mixture was placed in a capillary tube and attached by a rubber adapter to a No. 27 hypodermic needle that was inserted into the injection port of the

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gas chromatograph. The exchange reaction was done for 30 sec at 250°C by heating with a small resistance wire heater surrounding the capillary tube (Stephen and Teszler 1960). The carbonyl compounds were quantitated by comparing the integrated peak areas to those of the DNPH derivatives of authentic carbonyl compounds. Unknown peaks were quantitated against the nearest standard peak.

Furfural and hydroxymethylfurfural (HMF) were determined as HMF equivalent by extraction from the crust and crumb with benzene, and spectrophotometric measurement of the color after reaction with *p*-aminodimethylaniline stannous chloride (El Dash and Johnson 1970).

Flavor Evaluation

The multiple comparison difference test (Larmond 1970) was used for aroma and taste evaluations. Ten panelists were presented with 2% dispersions of protein supplements or slices of supplemented bread for evaluation of aroma and taste preferences. Bland 50 soy flour and bread containing 15% Bland 50 soy flour were used as the reference (R). The ratings were given numerical values with "no difference" equaling 5, "extremely better than R" equaling 9, and "extremely inferior to R" equaling 1. Analysis of variance and Duncan's multiple range test were conducted on an IBM-158 computer following the Kim and Kohout (1975) procedures.

TABLE I
Regeneration Efficiency of Standards and Composition of Carbonyl Compounds in Wheat Flour and Legume and Oilseed Protein Supplements

Peak No.	Carbonyl Compound	% Regenerated by Flash-Exchange GLC	Carbonyl Composition in mg/100 g on Dry Basis				
			Wheat Flour	Soy Flour	Sunflower Concentrate	Fababean Concentrate	Field Pea Concentrate
1	Ethanal	99.8	57 ± 3[a]	31 ± 1	55 ± 9	72 ± 14	44 ± 10
2	Unknown-1		16 ± 2		25 ± 1		13 ± 2
3	Propanal	93.8	11 ± 2	23 ± 1	10 ± 0	27 ± 2	17 ± 2
4	2-Propanone	81.0	378 ± 4	253 ± 13	398 ± 27	349 ± 60	387 ± 34
5	Butanal	80.0	9 ± 1	19 ± 3	13 ± 1	12 ± 1	10 ± 1
6	2-Butanone	87.1	128 ± 12	42 ± 6	103 ± 13	61 ± 3	111 ± 8
7	3-Methylbutanal	77.9	85 ± 8		245 ± 17	116 ± 24	146 ± 7
8	Unknown-2		18 ± 2	17 ± 2	13 ± 1	15 ± 2	24 ± 5
9	Pentanal	78.9	30 ± 2	65 ± 8	59 ± 8	17 ± 3	38 ± 1
10	Hexanal	69.9	154 ± 10	543 ± 58	193 ± 20	256 ± 35	228 ± 10
	Total		886	993	1,114	925	1,018

[a]Standard deviation (n = 2).

TABLE II
Carbonyl Compound Composition of All-wheat and Composite Flour Fermented Doughs[a]

Peak No.	Carbonyl Compound	Wheat	Soy	Sunflower	Fababean	Field Pea
1	Ethanal	915 ± 9[b]	562 ± 19	842 ± 25	2,189 ± 118	2,145 ± 193
2	Unknown-1	29 ± 2	38 ± 5	29 ± 4		
3	Propanal	17 ± 2	27 ± 3	17 ± 4	42 ± 2	54 ± 7
4	2-Propanone	574 ± 49	701 ± 46	661 ± 13	688 ± 33	564 ± 32
5	Butanal	16 ± 2	16 ± 3	19 ± 3	16 ± 2	21 ± 2
6	2-Butanone	119 ± 15	91 ± 12	84 ± 5	141 ± 21	90 ± 6
7	3-Methylbutanal	106 ± 19	248 ± 28	153 ± 18	187 ± 13	346 ± 33
8	Unknown-2	12 ± 1	37 ± 4	22 ± 4	106 ± 14	208 ± 17
9	Pentanal	20 ± 5	12 ± 1	17 ± 2	23 ± 4	49 ± 2
10	Hexanal	162 ± 32	357 ± 20	175 ± 2	319 ± 47	483 ± 31
	Total	1,970	2.089	2,019	3,711	3,960

[a]mg/100 g on dry basis.
[b]Standard deviation (n = 2).

TABLE III
Carbonyl Compound Composition of All-Wheat and Supplemented Bread Crumbs[a]

Peak No.	Carbonyl Compound	Wheat	Soy	Sunflower	Fababean	Field Pea
1	Ethanal	166 ± 3[b]	325 ± 30	252 ± 7	339 ± 14	457 ± 31
2	Unknown-1	37 ± 3	60 ± 10	75 ± 7	47 ± 7	59 ± 6
3	Propanal	25 ± 2	56 ± 8	24 ± 4	24 ± 3	37 ± 3
4	2-Propanone	982 ± 7	1,332 ± 40	1,152 ± 19	1,062 ± 24	1,155 ± 39
5	Butanal	25 ± 5	38 ± 7	25 ± 2	32 ± 3	37 ± 1
6	2-Butanone	306 ± 36	358 ± 15	397 ± 31	346 ± 45	300 ± 21
7	3-Methylbutanal	107 ± 18	840 ± 38	331 ± 27	341 ± 33	616 ± 42
8	Unknown-2	47 ± 11	151 ± 10	118 ± 5	464 ± 17	599 ± 40
9	Pentanal	83 ± 11	123 ± 11	95 ± 6	79 ± 3	59 ± 4
10	Hexanal	139 ± 7	1,096 ± 134	368 ± 13	375 ± 41	520 ± 32
	Total	1,917	4,379	2,837	3,109	3,839
	Furfural and hydroxymethylfurfural[c]	600 ± 90	1,470 ± 180	980 ± 230	1,230 ± 160	1,730 ± 180

[a]mg/100 g on dry basis.
[b]Standard deviation (n = 2).
[c]Spectrophotometric measurement.

RESULTS AND DISCUSSION

The carbonyl compounds in bread are frequently determined by regeneration from DNPH derivatives and direct injection into the gas-liquid chromatographic instrument (El Dash and Johnson 1970, Maga 1974). Incomplete regeneration, especially of the long-chain carbonyls, and decomposition of the carbonyls during flashing have been serious limitations of the technique. In the present study, the use of DMAB-oxalic acid as exchange reagent resulted in recoveries of about 70 to 100% (Table I). The corresponding values for the exchange reagent, α-ketoglutaric acid, which was used in previous studies (El Dash and Johnson 1970, Lorenz and Maga 1972b), ranged from 35 to 81%. As can be seen in Fig. 1, an effective separation of the regenerated carbonyl compounds was achieved by the DMAB-oxalic acid flash-exchange technique.

The protein supplements contained the same carbonyl compounds as wheat flour, except that soy flour contained no 3-methylbutanal and no unknown-1 (Table I). Fababean concentrate also contained no unknown-1. The principal carbonyl compounds were tentatively identified as 2-propanone, hexanal, 2-butanone, and 3-methylbutanal. The high levels of hexanal and 2-propanone in soy flour have been reported previously (Fujimaki et al 1965, Sessa et al 1969), the hexanal occurring as a result of lipoxygenase activity (Grosch and Schwencke 1969).

Ethanal became a major carbonyl compound in the yeast-fermented doughs but the soy-supplemented dough contained much less than the fababean and field pea doughs (Table II). The levels of 2-propanone, hexanal, 3-methylbutanal, and 2-butanone were also high in all doughs including the all-wheat and soy-supplemented doughs. The total volatile carbonyl contents in the fermented doughs were much greater than in the original flours and protein concentrates, especially in fababean- and field pea-supplemented doughs.

The bread crumbs and crusts were lower in ethanal contents than the doughs but the contents of other GLC-determined carbonyl compounds were generally much higher, especially in the crusts (Tables III and IV). While the total carbonyl content of the soy dough was intermediate, the levels in the crumb were comparatively high. However, total carbonyl contents (GLC + HMF) in dough and crusts were in the order of field pea > fababean > soy > sunflower > wheat. 2-Propanone, hexanal, 3-methylbutanal, ethanal, and 2-butanone were again major carbonyls in the crumb and crust. Unknown compound-2 occurred in higher concentrations in the supplemented breads than in the doughs, especially those containing legume proteins. The major carbonyl compounds in the breads were furfural and HMF, which occurred in particularly high concentrations in the legume-

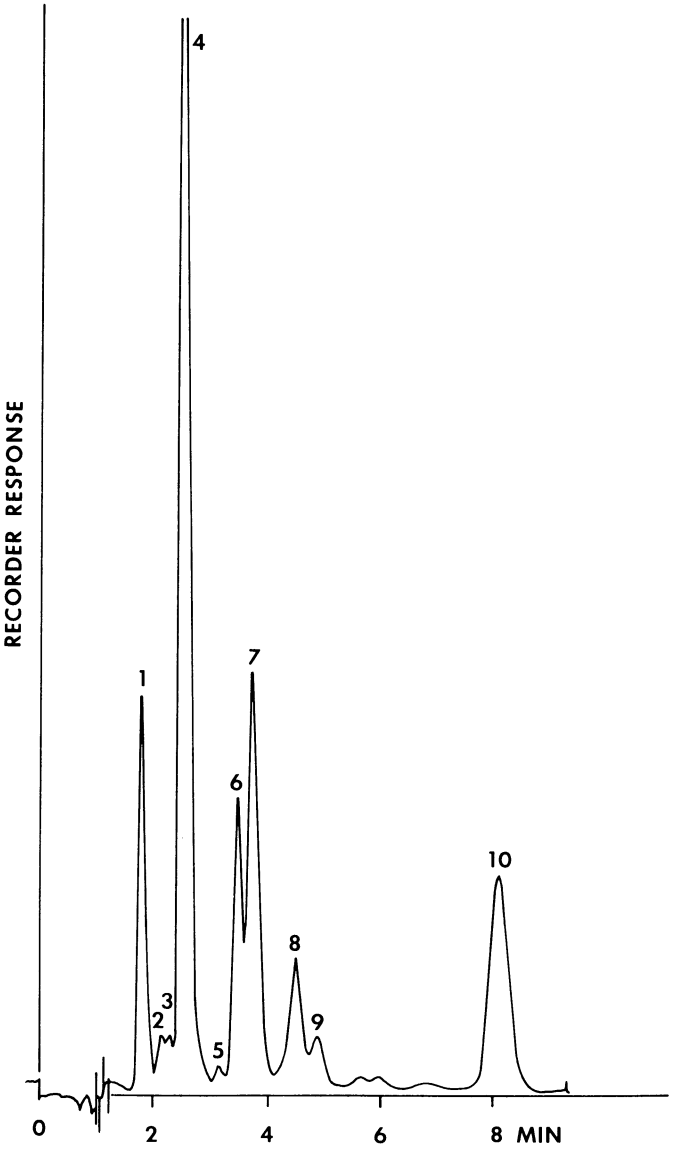

Fig. 1. Typical gas-liquid chromatogram of carbonyl compounds from fababean bread crust. Tentative identification: 1, ethanal; 2, unknown-1; 3, propanal; 4, 2-propanone; 5, butanal; 6, 2-butanone; 7, 3-methylbutanal; 8, unknown-2; 9, pentanal; 10, hexanal.

TABLE IV
Carbonyl Compound Composition of All-Wheat and Supplemented Bread Crusts[a]

Peak No.	Carbonyl Compound	Wheat	Soy	Sunflower	Fababean	Field Pea
1	Ethanal	149 ± 13[b]	410 ± 38	355 ± 14	331 ± 17	552 ± 48
2	Unknown-1	111 ± 6	45 ± 6	20 ± 1	87 ± 2	114 ± 14
3	Propanal	34 ± 2	62 ± 8	122 ± 5	77 ± 5	68 ± 9
4	2-Propanone	1,365 ± 30	1,893 ± 134	1,592 ± 61	1,602 ± 64	1,999 ± 250
5	Butanal	49 ± 3	68 ± 5	54 ± 8	63 ± 10	58 ± 5
6	2-Butanone	486 ± 26	656 ± 37	422 ± 20	420 ± 9	567 ± 47
7	3-Methylbutanal	237 ± 19	886 ± 60	468 ± 26	758 ± 56	826 ± 39
8	Unknown-2	70 ± 6	183 ± 20	87 ± 5	337 ± 18	418 ± 60
9	Pentanal	93 ± 13	228 ± 12	159 ± 26	124 ± 13	179 ± 23
10	Hexanal	419 ± 26	1,121 ± 61	489 ± 11	792 ± 72	728 ± 18
	Total	3,013	5,552	3,768	4,591	5,509
	Furfural and hydroxymethylfurfural[c]	2,920 ± 260	4,360 ± 280	2,470 ± 180	6,110 ± 920	5,760 ± 460

[a] mg/100 g on dry basis.
[b] Standard deviation (n = 2).
[c] Spectrophotometric measurement.

TABLE V
Flavor of Protein Supplements and Supplemented Breads
(1 = poor flavor, 9 = excellent flavor)

	Protein Supplement[a]		Supplemented Bread[a]	
Protein Supplement	Aroma	Taste	Aroma	Taste
Wheat flour control	...	...	6.6a	5.9a
Sunflower concentrate	4.4ab	4.5a	5.3b	4.2b
Heated soy flour	4.8a	2.9b	3.1c	4.4b
Fababean concentrate	4.1ab	2.9b	3.5c	4.2b
Field pea concentrate	3.6b	1.6c	2.1c	2.2c
Bland 50 soy flour	5.0a	5.0a	5.0b	5.0b

[a]Mean scores with the same letter are not significantly different at the 5% level.

supplemented breads containing soy, fababean, or field pea products. Furfural and HMF were determined in bread since they are known to be formed during baking and have specific effects on bread flavor (El Dash and Johnson 1970).

The high levels of some carbonyl compounds in supplemented breads could be attributed, in part, to the carbonyls present in the legume and oilseed protein supplements (Table I). Yeast fermentation increased the concentrations of ethanal and 2-propanone in all fermented doughs, whereas 3-methylbutanal and hexanal increased in some supplemented doughs (Table II). These changes in carbonyl contents have been reported to occur during normal fermentation of bread doughs (Kohn et al 1961). Losses of ethanal during baking (Tables III and IV) were expected although ethanal is also produced during Maillard-type reactions in the crusts (Rooney et al 1967). Nonenzymatic browning (El Dash and Johnson 1970, Linko et al 1962, Rooney et al 1967) resulted in the production of the other volatile carbonyl compounds such as hexanal, propanone, butanone, methylbutanal, furan compounds, and their derivatives, which were found to increase during bread making of supplemented flours in the present study, especially in the crusts. Furfural and HMF are produced during the Maillard reaction (Linko et al 1962, Rooney et al 1967) so that levels of both free amino acids and reducing sugars could affect the quantities of these volatile bread components.

The multiple comparison taste panel rated the legume and oilseed protein-supplemented breads as being lower than the all-wheat control in aroma and taste preferences (Table V). Sunflower and Bland 50 soy supplements and breads were assigned significantly higher aroma and taste scores than field pea; soy and fababean received intermediate sensory ratings. In protein supplement taste and bread aroma, the preference ratings were in the order of wheat > sunflower > soy = fababean > field pea. These flavor ratings were generally in the order of increasing total carbonyl contents of the protein-supplemented breads except for the particularly high level of carbonyl compounds in the soy bread crumb. The taste threshold values for carbonyl compounds were generally quite low (Borovikova et al 1971). Earlier Lorenz and Maga (1972a) found that aroma and taste acceptability decreased as the total carbonyl content increased with aging of bread. Recently, Rao et al (1978) found a significant inverse correlation between consumer preference and total carbonyl contents in breads prepared from wheat flour that had been treated with gamma-radiation.

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