Effect of Selected Oil and Essence Volatile Components on Flavor Quality of Pumpout Orange Juice

Esam M. Ahmed,* Raymond A. Dennison, and Philip E. Shaw

Selected volatile orange oil and essence components were added to pumpout juice at concentrations similar to those found in orange juice. Flavor quality of the modified pumpout juice was determined by three methods of sensory panels: trained, expert, and untrained. Pumpout juice containing acetaldehyde, citral, ethyl butyrate, *d*-limonene, and octanal received the highest score by the three methods of sensory panel used.

The purpose in identifying and quantifying volatile components in orange juice is for defining their contributions to the juice flavor quality. Patton and Josephson (1957) reported that the flavor significance of an individual compound may be determined by contrasting its concentration in a food with its threshold value in pure water. If the compound was present in excess of its threshold, it probably had a direct effect on flavor. Ahmed et al. (1978a) found that certain volatile constituents of orange juice, present in amounts exceeding their flavor threshold values in water, contributed to orange flavor. Volatile compounds present in orange juice in amounts below their threshold values in water contributed indirectly to orange flavor through additive or synergistic effects with other components (Ahmed et al., 1978a). Interactions among mixtures of basic taste components (Fabian and Blum, 1943; Keith and Powers, 1968; Kamen et al., 1961; Heinreiner, 1955a,b; Siek et al., 1969) on taste acuity of panelists were dependent on component concentration, solvent used, method of sample preservation, and number of panelists. These factors need to be considered or standardized in order to ascertain the relative contribution of a component to the flavor of a food. Volatile and nonvolatile components present in orange juice may interact with each other and influence its flavor (Ahmed et al., 1978b). Sugar and citric acid are major contributors to orange juice flavor and impart sweet and sour tastes to the juice (Tressler et al., 1939). Apparent flavor intensity of orange juice is achieved at a soluble solids and titratable acidity ratio of 15:1 and the presence of certain volatile components (Morse, 1954). A mixture of components in the proper proportions is believed responsible for the primary flavor of orange juice (Shaw, 1977), but there have been no studies reported that have shown the influence on flavor of individual or mixtures of known orange juice volatile components.

The purpose of the present study was to determine the relative contribution of individual and mixtures of volatile orange components to the flavor of orange juice.

EXPERIMENTAL SECTION

Materials. Two types of frozen orange juice concentrate (FOJC) were obtained from commercial sources known for their consistent production of high-quality FOJC. One type was the pumpout juice produced by the evaporative concentration process with no added oil, essence, or other flavor fractions. The other type was conventional FOJC prepared from the same pumpout juice plus added flavor components. Both types were used as reference juices in this study. The concentrated juices were diluted with distilled water (1:3 w/w) prior to either the addition of volatile components or presentation to the panelists. Acidities of both pumpout and reference juices were adjusted by the addition of citric acid to obtain Brix–acid ratios close to 15:1; the B/A ratio ranged from 14.8:1 to 15.4:1. Color and consistency of the pumpout and reference juices were similar. Orange volatile components were purchased from commercial sources and their purities were determined (Ahmed et al., 1978a) prior to use in flavor tests. Individual or mixtures of components were added to pumpout juice, just prior to sample presentation to the panelists, at concentrations normally found in orange juice (Table I). The pumpout juice with added components is referred to as modified pumpout juice.

Table I. Concentrations (ppb) of Selected Compounds Added to Pumpout Juice

Compound	ppb in juice	Reference
Acetaldehyde	3000	Kirchner, Miller (1957)
Citral	780	Shaw[a]
Citronellal	140	Shaw
Decanal	720	Shaw
Dodecanal	120	Shaw
Ethyl butyrate	400	
d-Limonene	190200	Shaw
Linalool	840	Shaw
Myrcene	5300	Shaw
Nonanal	10	Stanley et al. (1961)
Octanal	60	Lifshitz et al. (1970)
α-Pinene	1600	Shaw
trans-2-Hexenal	9000	Shaw

[a] Calculated from the data of Shaw and Coleman (1974) assuming a level of 0.02% oil present in the juice.

Sample Preparation. Orange juice samples were placed in 112-cm^3 odorless and tasteless plastic cups fitted with tight lids. Odorless ink was used to mark the cups with three-digit codes with the exception of reference juice samples which were marked R. Orange juice samples were served to the panelists slightly chilled at temperature 10 ± 2 °C (Larmond, 1973).

Sensory Testing. Flavor of pumpout, modified pumpout, and reference juices was evaluated by sensory panels. Sensory tests were conducted in duplicates for each treatment with the exception of the expert panel testing where it was carried out only once. Panelists were asked to take a deep sniff as soon as the cup lid was removed and then take a sip of the juice and swirl it in their mouth for a few seconds before swallowing. Panelists were requested to base their evaluation on the combined sensations of odor

Department of Food Science and Human Nutrition, University of Florida, Gainesville, Florida 32611 (E.M.A., R.A.D) and U.S. Citrus and Subtropical Product Laboratory, Southern Region, Agriculture Research Service, U.S. Department of Agriculture, Winter Haven, Florida 33880 (P.E.S).

Table II. Mean Sensory Scores[a] of Modified Pumpout Orange Juice in Comparison to Good Quality Reference Juice

Compound	Rating: Actual	Rating: Adjusted[b]	Rating: % of maximum
Acetaldehyde	3.9 ± 0.5	4.6	66
α-Pinene	4.1 ± 0.6	4.8	68
Citral	4.0 ± 0.4	4.7	67
Citronellal	4.0 ± 0.6	4.7	67
Dodecanal	4.0 ± 0.5	4.7	67
d-Limonene	3.5 ± 0.5	4.1	58
Ethyl butyrate	4.1 ± 0.8	4.8	68
Linalool	3.8 ± 0.6	4.4	63
Myrcene	2.8 ± 0.5	3.3	47
Nonanal	3.9 ± 0.4	4.6	66
Octanal	4.0 ± 0.3	4.7	67
trans-2-Hexenal (9000 ppb)	1.7 ± 0.3	2.0	28
(4500 ppb)	1.7 ± 0.3	2.0	28
(3000 ppb)	2.3 ± 0.4	2.7	39
Acetaldehyde and citral	2.4 ± 0.6	2.8	40
Acetaldehyde and dodecanal	4.5 ± 0.5	5.2	74
Acetaldehyde and ethyl butyrate	3.7 ± 0.5	4.3	61
Acetaldehyde and limonene	4.0 ± 0.4	4.7	67
Acetaldehyde and linalool	4.3 ± 0.4	5.0	71
Acetaldehyde and octanal	4.2 ± 0.4	4.9	70
Acetaldehyde and α-pinene	4.4 ± 0.5	5.1	73
Ethyl butyrate and linalool	4.5 ± 0.4	5.2	74
Ethyl butyrate and α-pinene	4.1 ± 0.5	4.8	68
Limonene and α-pinene	3.6 ± 0.6	4.2	60
Limonene and octanal	3.3 ± 0.5	3.8	54
Linalool and octanal	4.4 ± 0.5	5.1	73
Citral and dodecanal	4.1 ± 0.6	4.8	68
Citral and ethyl butyrate	4.6 ± 0.4	5.4	77
Citral and octanal	4.2 ± 0.4	4.9	70
Citral and α-pinene	4.1 ± 0.6	4.8	68
trans-2-Hexenal (3000 ppb) and acetaldehyde	1.7 ± 0.7	2.0	28
trans-2-Hexenal (3000 ppb) and ethyl butyrate	1.9 ± 0.5	2.2	31
trans-2-Hexenal (3000 ppb) and limonene	2.2 ± 0.4	2.6	37
trans-2-Hexenal (3000 ppb) and linalool	1.6 ± 0.6	1.9	27
trans-2-Hexenal (3000 ppb) and octanal	1.8 ± 0.6	2.1	30
α-Pinene, citral, ethyl butyrate, acetaldehyde	4.6 ± 0.5	5.4	77
Limonene, α-pinene, octanal	4.0 ± 0.4	4.7	67
Limonene, citral, ethyl butyrate, acetaldehyde, α-pinene	4.3 ± 0.4	5.0	71
Limonene, acetaldehyde, ethyl butyrate, octanal	4.6 ± 0.4	5.4	77
Limonene, acetaldehyde, octanal	4.2 ± 0.3	4.9	70
Limonene, ethyl butyrate, octanal	4.5 ± 0.4	5.2	74
Limonene, citral, acetaldehyde	5.1 ± 0.4	6.0	86
Limonene, citral, ethyl butyrate	5.2 ± 0.3	6.1	87
Limonene, citral, ethyl butyrate, acetaldehyde	5.0 ± 0.3	5.8	83
Pumpout orange juice	4.1 ± 0.4	4.8	68
Reference orange juice	6.0 ± 0.1	7.0	100

[a] Sensory scores: 7 similar to reference juice, 1 dissimilar to reference juice. Actual data presented as mean ± standard deviation. [b] Adjustment factor 1.1667.

and flavor by the mouth. Three types of panels were used:

1. Trained Panel. Panelists were selected on the basis of their performance in previous panels (Ahmed et al., 1978a,b). The selection was based on the consistency of results among replications of previous panels, ability to discriminate differences, having normal acuity, and dependability to attend panel sessions (Martin, 1973; Prell, 1976). Eleven judges (six males and five females ranging in age from 25 to 55 years of age evaluated the flavor of juice samples according to two different methods of scalar difference from control(s) (Mahoney et al., 1957; Siek et al., 1969):

A. Three modified pumpout orange juice samples and a reference juice sample were presented to the panelists. Panelists were asked to compare its odor and flavor to the reference juice and present their scores on a scale ranging from 7 for exactly like reference juice to 1 for extremely dissimilar to reference juice. Pumpout and reference juices were occasionally introduced as unknown to the panelists. There were 22 observations ($N = 22$) for each sample. Scores assigned to the reference juice (as unknown sample) were used to adjust modified pumpout sample scores on the basis that reference juice should have received the maximum score of 7. The adjusted scores were calculated as percents of the maximum score of 7.

B. Three modified pumpout samples and two reference juices were presented to panelists. The two reference juices consisted of a pumpout juice assigned a rating of 1 and a good quality juice assigned a rating of 10. Panelists were asked to smell and taste each sample and assign a score for its flavor as compared to the two known reference samples. The reference juices were presented occasionally as unknown samples. There were 22 observations ($N = 22$) for each sample. Scores were adjusted as mentioned in 1A.

2. Expert Panel. The expert panel was selected from persons who have had a great amount of experience in sensory evaluation of orange juice. Twelve of the panel members were employees of the U.S. Department of Agriculture Citrus and Subtropical Products Laboratory, Winter Haven, Fla. and 14 of the members were employees of the University of Florida Research and Education

Table III. Mean Sensory Flavor Scores[a] of Modified Pumpout Orange Juice in Comparison to Two References Representing Extremes of Flavor Quality

	Rating		
Compounds	Actual	Adjusted[b]	% of maximum
Pumpout orange juice	1.5 ± 0.5	2.0	20
Commercial orange juice	7.6 ± 0.1	10.0	100
Acetaldehyde, citral, *d*-limonene	5.0 ± 0.4	6.6	66
Acetaldehyde, citral, linalool	5.1 ± 0.4	6.7	67
Acetaldehyde, citral, octanal	3.9 ± 0.4	5.2	52
Acetaldehyde, citral, α-pinene	4.0 ± 0.3	5.3	53
Acetaldehyde, citral, *d*-limonene, α-pinene	4.7 ± 0.4	6.2	62
Acetaldehyde, citral, ethyl butyrate, *d*-limonene	4.3 ± 0.4	5.7	57
Acetaldehyde, ethyl butyrate, *d*-limonene, octanal	5.4 ± 0.4	7.3	71
Acetaldehyde, *d*-limonene, octanal	5.1 ± 0.3	6.7	67
Acetaldehyde, ethyl butyrate, citral, *d*-limonene, octanal	6.1 ± 0.2	8.1	81
Acetaldehyde, citral, citronellal, ethyl butyrate, *d*-limonene, octanal	4.2 ± 0.4	5.5	55
Acetaldehyde, citral, citronellal, ethyl butyrate, *d*-limonene, linalool, octanal, α-pinene	5.7 ± 0.3	7.5	75
Acetaldehyde, citral, citronellal, decanal, ethyl butyrate, *d*-limonene, linalool, myrcene, nonnanal, octanal, α-pinene, *trans*-2-hexenal	1.1 ± 0.1	1.5	15
Acetaldehyde, citronellal, *d*-limonene, linalool, α-pinene	5.3 ± 0.4	7.0	70
Acetaldehyde, citronellal, α-pinene	4.4 ± 0.4	5.8	58
Acetaldehyde, citral, citronellal, decanal, ethyl butyrate, *d*-limonene, linalool, octanal, α-pinene	3.4 ± 0.4	4.5	45
Acetaldehyde, decanal, *d*-limonene	4.2 ± 0.4	5.5	55
Acetaldehyde, citral, decanal	4.0 ± 0.4	5.3	53
Acetaldehyde, *d*-limonene, α-pinene	5.5 ± 0.3	7.3	73
Acetaldehyde, citral, ethyl butyrate	5.7 ± 0.3	7.5	75
Acetaldehyde, *d*-limonene, nonanal	4.9 ± 0.4	6.5	65
Acetaldehyde, *d*-limonene, linalool	5.2 ± 0.3	6.2	62
Acetaldehyde, citronellal, *d*-limonene	4.8 ± 0.4	6.3	63
Acetaldehyde, citral, citronellal	4.6 ± 0.4	6.1	61
Acetaldehyde, citral, ethyl butyrate, *d*-limonene	6.1 ± 0.2	8.1	81
Citral, ethyl butyrate, *d*-limonene	6.0 ± 0.2	7.9	79
Citral, *d*-limonene, α-pinene	4.8 ± 0.4	6.3	63
Citral, *d*-limonene, octanal	5.3 ± 0.3	7.0	70
Citral, *d*-limonene, citronellal	4.8 ± 0.4	6.3	63
Citral, ethyl butyrate, octanal	4.8 ± 0.3	6.3	63
Citral, ethyl butyrate, *d*-limonene, octanal, α-pinene	5.0 ± 0.4	6.6	66
Citral, linalool, octanal	4.9 ± 0.4	6.5	65
Citral, citronellal, ethyl butyrate	5.5 ± 0.4	7.3	73
Citral, *d*-limonene, nonanal	5.1 ± 0.3	6.7	67
Citral, decanal, *d*-limonene	3.5 ± 0.5	4.6	46
Citral, octanal, α-pinene	4.8 ± 0.4	6.3	63
Citral, *d*-limonene, linalool	5.7 ± 0.4	7.5	75
Citral, citronellal, octanal	4.6 ± 0.4	6.1	61
Citral, decanal, linalool	5.4 ± 0.4	7.1	71
Citral, ethyl butyrate, linalool	5.8 ± 0.3	7.7	77
Citral, ethyl butyrate, α-pinene	5.5 ± 0.3	7.3	73
Citral, ethyl butyrate, *d*-limonene, linalool	5.7 ± 0.4	7.5	75
Citral, ethyl butyrate, *d*-limonene, octanal	6.2 ± 0.3	8.2	82
Citral, ethyl butyrate, *d*-limonene, α-pinene	5.0 ± 0.3	6.6	66
Citral, citronellal, ethyl butyrate, *d*-limonene	5.8 ± 0.4	7.7	77
Ethyl butyrate, *d*-limonene, octanal	5.5 ± 0.3	7.3	73
Ethyl butyrate, citronellal, *d*-limonene	5.1 ± 0.4	6.7	67
Ethyl butyrate, *d*-limonene, linalool	5.8 ± 0.4	7.7	77
Ethyl butyrate, *d*-limonene, decanal	3.8 ± 0.5	5.0	50
Ethyl butyrate, *d*-limonene, nonanal	6.3 ± 0.5	8.3	83
d-Limonene, citronellal, linalool	4.7 ± 0.4	6.2	62
d-Limonene, decanal, α-pinene	3.1 ± 0.4	4.1	41
d-Limonene, linalool, α-pinene	5.1 ± 0.5	6.7	67
d-Limonene, decanal, linalool	3.4 ± 0.4	4.5	45
d-Limonene, nonanal, α-pinene	5.1 ± 0.5	6.7	67
d-Limonene, citronellal, linalool, octanal	5.1 ± 0.5	6.7	67
d-Limonene, citronellal, linalool, α-pinene	4.9 ± 0.5	6.5	65

[a] Sensory scores: 1 similar to pumpout juice, 10 similar to reference juice. Actual data presented as mean ± standard deviation. [b] Adjustment factor 1.321.

Center at Lake Alfred, Fla. Methods of sample presentation was similar to that of method B of the trained panel study. There were 26 observations ($N = 26$) for each sample.

3. *Untrained Panel.* One hundred and five untrained panelists ranging in age from 20 to 60 years and representing different sexes and races were screened for their ability to indicate if the flavor of pumpout juice containing *d*-limonene or pumpout juice containing *d*-limonene, acetaldehyde, citral, and ethyl butyrate closely resembled the flavor of good quality reference orange juice. Sixty-two panelists were capable of choosing the latter juice over the

former. These were the selected panelists who participated in this panel. There were 124 observations for each sample ($N = 124$). Two samples of the modified pumpout juice and the reference juice were presented to the panelists. Panelists were asked to indicate which modified pumpout juice was similar in flavor to the reference juice. Results were expressed as percentage of panelists selecting any given modified pumpout juice. Statistical analysis were limited to calculations of the mean and standard deviation for each treatment.

RESULTS AND DISCUSSION

Preliminary investigations have shown that: (1) sensory ratings of orange volatile components added to pumpout juice were consistently higher than when volatile components were added to water; (2) the variations in the sensory ratings of the pumpout and reference juices at different times of presentation to the panelists were due to the variations in degrees Brix, acidity, and their ratios. On several occasions the panelists described the pumpout juice as sweeter, and sometimes as more sour, than the reference juice. Therefore, in this study the Brix–acid ratios of the pumpout and reference juices were adjusted as close to 15:1 as possible and pumpout juice was used as the carrier of the volatile components. In addition, the use of pumpout provided the panelists with samples similar to the reference juice in appearance, color and consistency.

1. Trained Panel. Sensory flavor scores of the pumpout, modified pumpout, and reference juices are shown in Table II. The following information is obtained from this table: (1) pumpout juice received an average score of 4.1, indicating a response of neither similar or dissimilar to reference juice to slightly similar to reference juice; (2) pumpout juice modified by the addition of most individual components and some of the two-component systems received lower scores than the pumpout juice; (3) *trans*-2-hexenal used alone or in combination with the other components resulted in juices receiving low scores; (4) the reference juice, introduced as unknown sample, received an average score of 6.0 (moderately similar to reference juice), and (5) pumpout juice containing two or more volatile component mixtures of acetaldehyde, α-pinene, citral, ethyl butyrate, *d*-limonene, linalool, or octanal received sensory scores ranging from 4.5 to 5.2 (slightly similar to moderately similar to reference juice).

It was felt that improvement in the precision of the sensory data could be achieved by presenting to the panelists, in addition to the modified pumpout samples, two references juices, one containing no volatile components (pumpout juice) and the other a good quality juice. In addition, the sensory score range was expanded from 1 for the pumpout to 10 for the good quality juice. The good quality juice was presented occasionally as unknown sample. Sensory scores were adjusted as mentioned previously. Panel responses are shown in Table III and could be summarized as follows: (1) good quality orange juice presented as unknown received the highest actual sensory score; (2) mixtures of compounds containing acetaldehyde, citral, ethyl butyrate, *d*-limonene, linalool, octanal, or α-pinene added to pumpout juice resulted in scores ranging from 70 to 83% of maximum score; (3) the highest scores were obtained for the mixtures of: acetaldehyde, citral, ethyl butyrate, and *d*-limonene (81%); citral, ethyl butyrate, and *d*-limonene (79%); citral, ethyl butyrate, *d*-limonene, and α-pinene (82%); and ethyl butyrate, *d*-limonene, and nonanal (83%); (4) addition of either decanal, citronellal, or *trans*-2-hexenal lowered the scores of the modified pumpout juice; and (5) the mixture containing *d*-limonene, linalool, and α-pinene received an adjusted score of 6.7. Substitution of α-pinene by decanal lowered the score to 4.5. The mixture containing citral, *d*-limonene, and ethyl butyrate received a score of 7.9. Substitution of ethyl butyrate by citronellal lowered the rating to 6.3. The lowest score was obtained as the mixture contained *trans*-2-hexenal.

2. Expert Panel. The highest percentages of maximum scores were assigned to pumpout juice containing acetaldehyde, citral, ethyl butyrate, *d*-limonene, and octanal (78 ± 4%); acetaldehyde, citral, ethyl butyrate, and *d*-limonene (75 ± 3%); and acetaldehyde, citral, ethyl butyrate, *d*-limonene, and α-pinene (70 ± 4%).

In addition, the expert panel at Lake Alfred rated the orange juice samples on a hedonic scale. Their mean rating scores for modified pumpout juices (6.2–6.8) were slightly less than the score for the good quality reference juice (7.2). A hedonic rating of 6 indicates like slightly, 7 indicates like moderately, and 8 indicates like very much.

3. Untrained Panel. Results of the untrained panel selection of modified pumpout orange juice indicated that samples receiving the highest percentages of selection were pumpout juice containing acetaldehyde, citral, ethyl butyrate, *d*-limonene, and octanal (92%); acetaldehyde, ethyl butyrate, *d*-limonene, and octanal (87%); and acetaldehyde, citral, ethyl butyrate, *d*-limonene, and α-pinene (82%).

It can be concluded that pumpout juice containing acetaldehyde, citral, ethyl butyrate, *d*-limonene, and octanal received the highest scores (78–92%) by the three methods of sensory panels used. Results obtained show that certain 3–5 component mixtures of orange juice volatiles can provide an acceptable orange flavor to pumpout juice. Further studies with other multicomponents are needed to better define the contribution of these components to orange juice flavor.

LITERATURE CITED

Ahmed, E. M., Dennison, R. A., Dougherty, R. H., Shaw, P. E., *J. Agric. Food Chem.* **26**, 000 (1978a).

Ahmed, E. M., Dennison, R. A., Dougherty, R. H., Shaw, P. E., *J. Agric. Food Chem.* **26**, 000 (1978b).

Berg, H. W., Filipello, F., Hinreiner, E., Webb, A. D., *Food Technol.* **9**, 23 (1955).

Fabian, F. W., Blum, H. B., *Food Res.* **8**, 179 (1943).

Heinreiner, E., Filipello, F., Webb, A. D., Berg, H. W., *Food Technol.* **9**, 351 (1955a).

Heinreiner, E., Filipello, F., Webb, A. D., Berg, H. W., *Food Technol.* **9** 489 (1955b).

Kamen, J. M., Pilgrim, F. J., Gutman, N. J., Kroll, B. J., *J. Exp. Psychol.* **62**, 348 (1961).

Keith, E. S., Powers, J. J., *J. Food Sci.*, **33**, 213 (1968).

Kirchner, J. G., Miller, J. M., *J. Agric. Food Chem.* **5**, 283 (1957).

Kramer, A., Murphy, E. F., Briant, A. M., Wang, M., Kirkpatrick, M. E., *J. Agric. Food Chem.* **9**, 224 (1961).

Langler, J. E., Day, E. A., *J. Dairy Sci.* **47**, 1291 (1964).

Larmond, E., *Food Technol.* **27**, 28(11) (1973).

Lifshitz, A., Stanley, W. L., Stepak, Y., *J. Food Sci.* **35**, 547 (1970).

Mahoney, C. H., Stier, H. L., Crosby, E. A., *Food Technol.* **11**(9), 29 (1957).

Martin, S., *Food Technol.* **27**, 22(11) (1973).

Meijboom, P. W., *J. Am. Oil Chem. Soc.* **41**, 326 (1964).

Morse, R. L. D., *Proc. Fla. State Hortic. Soc.* **67**, 186 (1954).

Pangborn, R. M., *Food Res.* **25**, 245 (1960).

Pangborn, R. M., *J. Food Sci.* **26**, 648 (1961).

Patton, S., *Josephson*, D. V., *Food Res.* **22**, 316 (1957).

Prell, P. A., *Food Technol.* **30**, 40(11) (1976).

Siek, T. J., Alhin, I. A., Sather, L. A., Lindsay, R. C., *J. Food Sci.* **34**, 265 (1969).

Shaw, P. E., in "Citrus Chemistry and Technology", Vol. 1, Nagg, S., Shaw, P. E., Veldhuis, M. K., Ed., Avi Publishing Co., Westport, Conn., 1977.

Shaw, P. E., Ahmed, E. M., Dennison, R. A., Proceedings of the International Society of Citriculture, "Orange Juice Flavor: Importance of Certain Volatile Components as Determined by Sensory Panel Evaluations", in press, 1977.
Shaw, P. E., Coleman, R. L., *J. Agric. Food Chem.* **22**, 785 (1974).
Stanley, W. L., Ikeda, R. M., Vannier, S. H., Rolle, L. A., *J. Food Sci.* **26**, 43 (1961).
Tressler, D. K., Joslyn, M. A., Marsh, G. L., "Fruit and Vegetable Juice", Ave Publishing Co., Westport, Conn., 1939.

Received for review July 21, 1977. Accepted November 18, 1977. Florida Agricultural Experiment Stations Journal Series No. 612. Mention of a brand name is for identification only and does not constitute endorsement by the U.S. Department of Agriculture over others which may also be suitable.

Volumetric Determination of Total Aldehydes in Citrus Oils

M. L. Shankaranarayana,* B. Raghavan, K. O. Abraham, and C. P. Natarajan

A titrimetric method for determining citral has been developed; it is based on the quantitative reaction of the citral with semicarbazide hydrochloride to form a semicarbazone. The semicarbazone is removed and the excess unreacted semicarbazide determined iodometrically using chloramine-T reagent. Two moles of chloramine-T are required per mole of semicarbazide. The method has been applied for the determination of the total aldehydes in citrus oils (expressed as citral in lemon and lime and as decanal in orange). The present method can be used in the routine quality evaluation of citrus oils, and the sample required is in the range of 0.5 to 1.0 g.

Citrus oils are a major by-product of the citrus processing industry. The commercially important peel oils include lime, lemon, orange, grapefruit, bergamot, etc. These are used as flavoring ingredients in a variety of foods like soft drinks, ice cream, frozen desserts, confectionery, baked goods, and also in perfumery industry. The oils chiefly consist of terpene hydrocarbons (80–95%) and oxygenated terpenes such as citral, decanal, nootkatone linalyl acetate, terpineol, geranyl acetate, etc. The hydrocarbons are unstable and susceptible to photochemical and oxidation reactions and slowly contribute to the deterioration in the quality of the oils. They act largely as the carriers for the oxygenated compounds which are mainly responsible for the characteristic citrus flavors. With a view to obtaining stable and concentrated citrus oils, they are either partly or completely deterpenated. This is achieved by employing a variety of techniques like solvent partition (Merory, 1968; Ruys, 1957), column chromatography (Anandaraman et al., 1976; Braverman and Solomiansky, 1957; Kirchner and Miller, 1952; Rovesti and Rovesti, 1967; van der Lijn and Lifshitz, 1969), fractional distillation, (Lifshitz et al., 1969), etc.

Among the various oxygenated constituents, the aldehydes have been considered to have the most profound influence on the flavor quality of the citrus oils. For example, the characteristic odor of lemon and lime is mainly due to citral. Several methods have been described in literature for the determination of the aldehyde(s) content. As early as 1909, Hiltner described a colorimetric method for the determination of α,β-unsaturated aldehydes using *m*-phenylenediamine reagent. The accuracy of the method has been improved by using the photoelectric spectrophotometer (Kleber, 1912). The volumetric method, employing phenylhydrazine reagent, was adopted as the USP procedure (1965) for sometime for the determination of citral in lemon oil. Stanley et al. (1958) described a specific method for the determination of citral in lemon oil based on its reaction with vanillin–piperidine reagent giving a green color. Levi and Laughton (1959) reported an ultraviolet absorption spectrophotometric method wherein the citral is converted into its barbituric acid derivative (λ_{max} 336 nm). The colorimetric method of Ismail and Wolford (1970) employs *N*-hydroxybenzenesulfonamide reagent which determines the total aldehydes. This method has undergone some modifications in the hands of Dougherty and Petrus (1971) and Petrus et al. (1970). Surve et al. (1958) have described an ultraviolet spectrophotometric method for citral based on its characteristic absorption peak around 238 nm. The most widely used method for the determination of the aldehydes in citrus oils is the volumetric procedure employing hydroxylamine hydrochloride reagent (Bennett and Salamon, 1927, 1930; AOAC, 1970).

During our work on the quality evaluation of citrus oils, it was necessary to determine the aldehyde content in comparatively small samples (0.5 to 1 g) of the citrus oils. Therefore, a titrimetric method has been developed using chloramine-T (sodium derivative of *N*-chloro-*p*-toluenesulfonamide) reagent. The aqueous solution of this reacts as if it were a hypochlorite (Bishop and Jennings, 1958):

$$RNCl{\cdot}Na + H_2O \rightarrow RNH_2 + NaOCl \quad (1)$$

where R = $CH_3C_6H_4SO_2$-. Chloramine-T has been recently employed for the determination of flavor strength in mustard (Shankaranarayana et al., 1972), asafetida (Abraham et al., 1973), and radish (Damodaran, 1975), and also for the determination of the derivatives of dithiocarbamic acid (Abraham et al., 1975). The present method consists of the quantitative conversion of the aldehydes into their semicarbazone derivatives (eq 2) and then determining the unreacted semicarbazide oxidimetrically using chloramine-T reagent (eq 3):

$$RCHO + NH_2CONHNH_2 \rightarrow RCH{=}NNHCONH_2 + H_2O \quad (2)$$

Central Food Technological Research Institute, Mysore-570013, India.

0021-8561/78/1426-0372$01.00/0