lary column unless otherwise indicated. The mass spectra of all compounds reported were in complete agreement with those of the known compounds. Many of the sulfur compounds identified in the model systems studied are naturally occurring. Some of the foods in which they occur are reported in Tables I and II. Of the 24 compounds listed in Table I, all but six have been reported as naturally occurring.

The sulfur compounds identified in the D-xylose–L-cysteine·HCl model system are given in Table II, along with their I_E values and natural occurrence. Those compounds designated by an asterisk were identified in both model systems.

SUMMARY

Two model systems approximating the conditions of cooked meats were prepared and analyzed. A total of 34 sulfur compounds were identified. Of these, 21 have been previously reported in natural foods, and the remaining 13 have not been reported as naturally occurring. Eleven of these compounds have been synthesized.

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Identification and Analysis of the Major Acids from Fruit Juices and Wines

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The major organic acids of fruit juices and wines have been investigated using lead precipitation and glc of the trimethylsilyl derivatives. The identity of individual acids was established by glc, thin-layer chromatography, and mass spectral data. The amounts of individual nonvolatile acids were calculated using an internal standard and standard curves. The major differences between juices and wines are the greater overall amounts of acids in the former and the presence of lactic and succinic acids in the latter. A comparison was made between the amounts of acids found by two different nonequivalent methods, glc and titration. In juices the two methods gave similar results but in wines the values were different. The phosphoric acid content of juices was analyzed *via* glc at the same time as major organic acids and the values were found to be comparable with those of a colorimetric method.

The acidic components of fruit juices and wines impart important properties to these foods being prominent in flavor, processing, and preservation. The acidity has also been used as a criterion of adulteration of one fruit with another. Because of these attributes the acids have been extensively studied in the past (Amerine and Cruess, 1960; Hulme, 1970; Tressler and Joslyn, 1961). These studies have employed mainly chemical and enzymatic methods in order to separate, identify, and quantify the acids. In recent years, with the advent of newer and less tedious chromatographic techniques, the acids have again been investigated by numerous workers (Chan *et al.*, 1972; Fernandez-Flores *et al.*, 1970; Martin *et al.*, 1971; Weissberger *et al.*, 1971). These analyses have allowed the major four to seven organic acids to be estimated and the amounts of individual acids to be determined quantitatively. Nevertheless, the identification of the acidic components from various fruit sources has been cursory and sometimes contradictory, often being based only on a single glc peak. In addition, the amounts of acids present in fruit juices and wines have received little attention when compared to the qualitative aspects. The comprehensive nature of the glc technique, whereby all acids are estimat-

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ed in one experiment, has also not been fully exploited. For these reasons, a study of the identity and amounts of acids in fruit products was initiated.

Brunelle *et al.* (1967) and again Martin *et al.* (1971) found wine to contain the acids succinic, tartaric, malic, and citric. Fernandez-Flores *et al.* (1970) reported most of the above acids in common fruits as well as significant (easily measurable) amounts of fumaric and glycolic acids. Markakis and Kallifidas (1971) investigated the organic acids of Concord grape wine and found a variety of common acidic components plus other minor types such as galacturonic and lactic acids; they also expressed their values found by ion exchange separation as a percent of total nonvolatile acidity. In a study of the components of apple juice, Ryan (1972) found malic, citric, quinic, and phosphoric acids and no others in significant amounts. Hence the various reports of constituents of fruit acids show the presence of malic, tartaric, and citric acids in many cases but the identity of these and of the other components, often present in smaller quantities, has not always been certain.

It is the purpose of this paper to identify the major organic acids from fruit juices and wines using a variety of techniques, including glc–ms. The amounts of acids present are determined by glc of their trimethylsilyl (TMS) derivatives and compared to the amounts as found by titration with base. A comparison is also made between the phosphoric acid content of fruits using the glc method and a colorimetric one.

METHOD

Samples. Fruit juices and fruit were obtained at local markets from the shelves. Apple juice was acquired by the Canada Department of Agriculture from inspected plants. Fruit wines were purchased at Government outlets. All samples were either analyzed immediately as purchased or stored at −10° after being opened.

Glc Analysis. *Apparatus.* A Hewlett-Packard 5750 gas–liquid chromatograph equipped with a flame ionization detector was used for the analysis with helium as the carrier gas (30 ml per min at the detector at room temperature). Stainless steel columns (6 ft × ⅛ in. o.d.) containing either 3% SE-30 or 3% OV-17 on acid-washed Chromosorb W (60–80 mesh) were used. The temperature program was 80–165° at 4° per min with a 3-min postinjection hold and an 8-min upper limit hold. The detector block and injection port were operated at 230°. Electrometer setting varied from 10^2–16 to 10^2–128, depending on the concentration of the acid being measured.

Reagents. Lead diacetate trihydrate (16 g) was dissolved in 100 ml of distilled water. Glutaric acid (1 mg/1 ml) solution in 95% ethanol was prepared daily.

Sample Preparation. Method A. For fruit juices with acid concentration greater than 15 mequiv/100 ml. These were analyzed essentially as in the method used by Fernandez-Flores *et al.* (1970). The alcohol content of the medium for lead salt precipitation was maintained at 80%. In some juices samples where the individual acid levels varied widely, it was necessary to run two different aliquots of varying strength with the same amount of glutaric acid in order to effect good quantification.

Method B. For all wines and fruit juices with low (less than 15 mequiv/100 ml) acid concentration. This method was identical to that of method A except for the following conditions. Only half as much sample, standard, and lead acetate solution was used, resulting in a final concentration of a least 90% alcohol. The depectinization and washing of lead salts were omitted since wine contains little pectins and sugars and derivatization was accomplished with exactly one-half the amount of reagents so that the final concentration of standard was the same as in method A.

Standards. These were treated as in methods A or B using their lead salts in the same concentration range as found in the sample. Graphs of the relative area of 1 mg of glutaric acid *vs.* weight of the acids were drawn and used to calculate the amount of acid in a sample.

Thin-Layer Chromatography. Tlc plates were coated with microcrystalline cellulose and spotted, eluted, and detected as described by Ryan (1972).

Glc–Mass Spectrum. A Perkin-Elmer Model 990 gas–liquid chromatograph with flame ionization detector with either 3% OV-17 or 3% SE-30 on Chromosorb W (60–80 mesh) acid-washed columns (*cf.* glc method) was coupled with an Hitachi RMS-4 Mass Spectrometer. The ionization voltage was 80 eV and source was maintained at 250°. Glc conditions were identical to those described in the Quantitative Method section, except that usually more than one injection of a sample was needed to investigate each peak of interest.

Titratable Acidity. *Simple Titratable Acidity (TA).* A 1–10-ml sample, depending on the amount of acid present, was diluted with about 150 ml of freshly boiled distilled water and titrated with 0.1 *N* NaOH using a glass electrode to a pH of 8.8. The end point was taken as the value where the pH had the maximum change with addition of base. This value, different for each fruit type, was between 7.2 and 8.4 pH units.

Total Titratable Acidity (TTA). A column (10 ml wet volume) of Dowex 50 WX12 strongly acidic cation exchange resin was purified by washing successively with alkali, acid, and water. The wine or juice (same sample size as in TA) was pipetted directly onto the column and acids were completely washed off with 30–50 ml of water. The sample was titrated as in TA. The column was changed for every fifth sample.

Phosphoric Acid. The phosphate present in juices and wine was measured by the molybdenum blue method both before and after digestion (inorganic and total phosphorus) as described by Vandercook (1969). Ascorbic acid was used as the reducing agent and the absorbance of blue complex was measured at 820 mμ. The only modification used was that, after the digestion step, the sample was neutralized to pH 8.2 with 0.1 *N* NaOH.

RESULTS AND DISCUSSION

All fruit juices and wines were analyzed for their principal acidic components by precipitation as insoluble lead salts. Nevertheless, complete precipitation of all acids is a problem. With fruit juices it was found that all the fixed or organic acids were deposited if the alcohol content of the media were 80%. Washing the precipitated salts with 80% aqueous ethanol resulted in little or no loss of acids with removal of the sugars which interfered on glc analysis. In the case of wines where both the total amounts of acids and sugars are lower, it was found that considerable losses of some acids (malic, glutaric, succinic) occurred unless the alcohol content of the precipitating media was increased to 90%. In addition, a coprecipitation phenomenon was involved in this method. If the total amounts of all acids were low in a sample, then complete precipitation of the acids did not occur unless either the water content was low or a foreign acid was added to sample. In the latter case all acids, including the ones in smaller quantities, then became insoluble.

Identity and Detection. *Glc.* The mixture of TMS derivatives of acids was resolved on both 3% SE-30 and 3% OV-17. A typical tracing from Bordeaux red wine is shown in Figure 1. Identity of all peaks was supported by injecting standard samples of the acid under study at the same time as the fruit sample and noting whether their retention times coincided. From the glc data alone, a good estimate of both the amount and kind of acidic component present could be obtained.

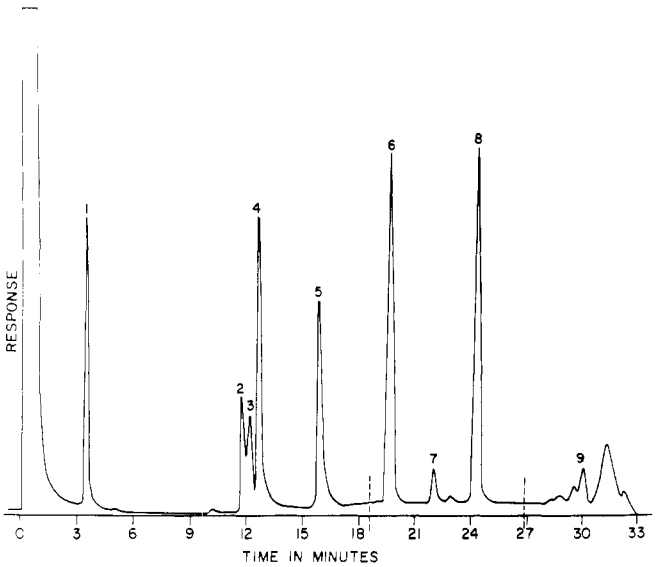

Figure 1. Glc chromatogram of organic acids from Bordeaux red wine. 1, lactic; 2, H_3PO_4; 3, unknown; 4, succinic; 5, glutaric (internal standard); 6, malic; 7, unknown; 8, tartaric; 9, sugars. Recorder range 10^2–16 except for peaks 6–8, which were 10^2–64.

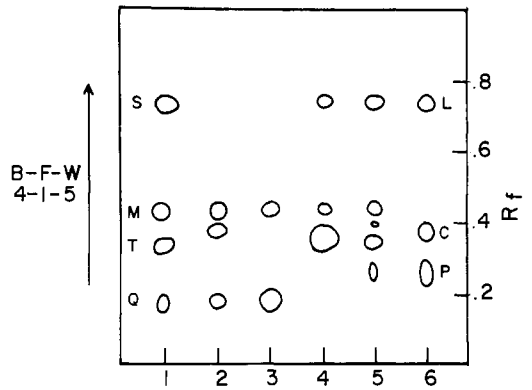

Figure 2. Tlc chromatogram of organic acids from regenerated lead salts. 1–6, standards (S, succinic; M, malic; T, tartaric; Q, quinic; L, lactic; C, citric; P, phosphoric); 2, cranberry juice; 3, prune juice; 4, strawberry wine; 5, Concord grape wine.

Tlc. As further support for the presence/absence of a particular acid, the acids were spotted on cellulose thin layers along with standard acids. R_f values of the acids have been reported by Chan *et al.* (1972). These tlc patterns were compared to those found by glc. Typical patterns of a number of samples are shown in Figure 2. All acids can be distinguished from each other with this technique as long as one is not present in excessive amounts.

Glc–Ms. Because of the sometimes conflicting reports of the occurrence of acids, it was deemed necessary to provide absolute identity of principal acids present in fruit samples. To this end TMS derivatives of acids from fruit sources and from standards were separated on a glc column whose effluent was fed directly into a mass spectrometer (ms). The virtue of this technique is that the coincidence of the molecular peak and cracking pattern from an unknown glc peak with that of a standard allows positive identification of the unknown. All of the acids were recorded as standards and each acid peak was analyzed from any source where there has been either a conflicting report or cursory identification. Two representative patterns of succinic and malic acids are shown in Figures 3 and 4. Other results are tabulated in Table I. Some of the salient features of this information are the following. All major organic or fixed acids found in fruit juices and wines are capable of being eluted from glc columns and determined *via* ms as their TMS derivatives. Each acid is completely silylated, *i.e.*, each position containing either an hydroxyl or carboxylic acid is silylated. Thus, besides verifying the type of acid from a particular source, the glc-ms procedure allows one to ascertain the degree of silylation of the derivative formed, an identity which could find use in other fields.

Quantitative Determination. In order to estimate the amounts of the principal acids in juices and wines, both an internal standard and standard curves were used. Graphs of the relative areas to glutaric acid *vs.* weight of the acids were determined using lead salts. Averages of duplicate injections of standards and samples were used. The values found are listed in Table II. It should be noted that some of the juices and wines are commercial products which can contain other ingredients such as citric acid and water for processing purposes. Thus, the total concentration of acids found in these samples does not always coincide with that found in the fruit. The well known predominance of malic in pome fruits, tartaric in grape products, and citric in citrus fruits is apparent. Succinic acid was found in only two juices in measurable amounts, either by glc or glc–ms. Neither fumaric nor glycolic acids could be detected as reported by Fernandez-Flores *et al.* (1970), even though added acid could be readily recovered from spiked samples. It would appear that these compounds, if present, are so in very small quantities (less than 5 mg/100 ml; limit of detection).

There are two significant differences between the juices and wines. The first is that all wines contain large amounts of both lactic and succinic acids, whereas in juices the former is lacking and the latter is found only in some samples (2 out of 18). This follows from the known fact (Amerine and Cruess, 1960) that these two acids are products of alcoholic fermentation and are not usually

Table I. Mass Spectra of Silylated Derivatives of Fruit Acids and Their Sources

Acid	Mol wt: Free acid	Mol wt: Fully silylated	Source[a]	*m/e* found[b]	Number TMS groups
Lactic	90	234	7, a–g	234m, 219s	2
Phosphoric	98	314	1, 5–6, 10, b–g	314m, 299s	3
Succinic	118	262	6–7, a–g	262m, 247s	2
Malic	134	350	1–6, 8–10, a, b, f, g	350m, 335m	3
Tartaric	150	438	10, a–d	438m, 423m	4
Citric	192	480	1, 3–9, e–g	480w, 374m	4
Quinic	192	552	2, 8	552w, 537s	5

[a] Sources. (i) Juices: 1, apple; 2, prune; 3, lemon; 4, lime; 5, pineapple; 6, strawberry; 7, raspberry; 8, cranberry; 9, black currant; 10, grape. (ii) Wines: a, Concord white; b, Concord red; c, Bordeaux white; d, Bordeaux red; e, apple; f, strawberry; g, red currant. [b] Strength of peaks; s, strong; m, medium; w, weak.

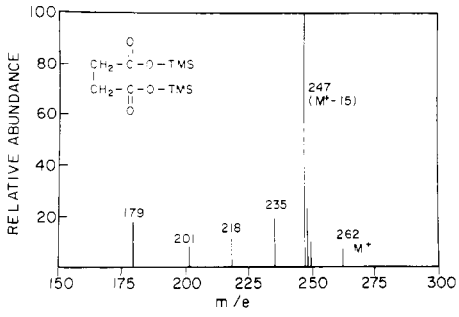

Figure 3. Mass spectral diagram of TMS derivative of succinic acid from strawberry wine.

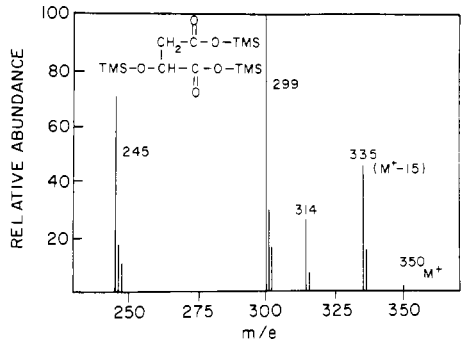

Figure 4. Mass spectral diagram of TMS derivative of malic acid from grape juice.

present in the juices. The second major difference between juices and wines is the smaller amounts of acids in wines as a result of fermentation and metabolic processes.

Method Comparison. The results as given in Table II show the amounts of acids determined as their insoluble lead salts, the fixed acids, and include free (H form) as well as cationic forms (salts). The volatile acidity is not measured by the glc method, as the lead salts of the volatile acids (formic and acetic) are soluble in the precipitating media. It was expected that some correlation would exist between the organic acids by lead precipitation and glc and the acids found by titration. Ideally, the so-called fixed or nonvolatile acids plus the volatile acidity should approximate the total titratable acidity (TTA), provided all fixed acids are precipitated as their lead salts and measured by glc. For this reason the titration of juice and wine samples was performed both before and after passage through a cation exchange column. The latter procedure converts all salts to the free acids and allows the calculation of the total titratable acidity (TTA) as well as the titratable acidity (TA) before column exchange. At the same time a measure is given of the amount of acids present in free and salt form. Such a comparison is made in Table III. It is found that in the case of apple, grape, and most of the fruit juices there is good correlation between organic acids by glc and the TTA, *i.e.*, the former is slightly smaller than the latter. This would indicate that the amount of volatile acidity is small and the glc method measures all major acidic components.

On the other hand, the closeness between the two methods with the fruit wines is not as good, the glc method being considerably lower. Similar results with wines (mediocre agreement) have been found by Martin *et al.* (1971) and with fruit (good agreement) by Chan *et al.* (1972). However, a difference between these two methods is to be expected in some cases and, in particular, with wines. These latter beverages contain other acidic components (higher volatile acidity, carbon dioxide, sulfates) in significant amounts which are not measured by lead precipitation-glc and which are measured by the TTA method. Hence, the difference between the two methods is attributed to the methods themselves which measure different things and not to a fault in amounts of individual acids found by lead precipitation-glc. In the latter method organic acids from spiked wines could be readily recovered. Moreover, Brunelle *et al.* (1967) found that the lead precipitation-glc method gave similar results to the AOAC fruit method for tartaric acid, supporting the contention that all major organic acids from wine are being measured by our method.

The glc-TMS method offers a simple way to identify and quantitize the individual major fixed or organic acids in a fruit sample by a single experiment, whereas many existing methods measure but one acid at a time. Another advantage over other procedures such as ion exchange and freeze-drying is that no evaporation step is involved and no losses occur as a result of volatility. The shortcomings

Table II. Amounts of Principal Acids from Fruit Sources, mg/100 ml

Sources	Lactic	H_3PO_4	Succinic	Malic	Tartaric	Citric	Quinic	Total, g/100 ml
Juices								
Apple[a]		19		640		14	28	0.70
Prune				171			620	0.79
Lemon				130		5630		5.76
Lime				100		7000		7.10
Pineapple		12		188		605		0.80
Strawberry		25	25	345		580		0.98
Raspberry	12		55			2480		2.54
Cranberry				180		202	250	0.63
Black currant				30		1170		1.20
Grape[b]		29		470	530			1.01
Wines								
Concord white	28		27	203	300			0.56
Concord red	206	33	59	190	175			0.66
Bordeaux white	221	9	50		180			0.46
Bordeaux red	225	5	68		73			0.37
Apple	192	8	43			200		0.44
Strawberry	28	7	75	25		570		0.71
Red currant	60	57	74	45		360		0.60

[a]Average of six samples. [b]Average of four samples. All other sources are single samples.

Table III. Amounts of Major Acids (mequiv/100 ml) in Juices and Wine by glc and by Titration

	Glc[a]	TTA[b]	TA[c]
Apple juice	10.3	10.6	6.9
	10.8	11.8	8.6
	9.9	10.5	7.4
	8.9	9.4	6.2
	11.0	10.8	7.2
	10.6	10.4	7.0
Grape juice	15.9	18.1	13.3
	14.0	16.1	11.9
	13.2	17.4	12.6
	14.1	17.9	12.5
Prune	5.8	15.1	7.5
Lemon	90.0	87.8	81.8
Lime	110.0	91.6	88.0
Pineapple	12.5	17.8	12.0
Strawberry	15.1	14.6	10.0
Raspberry	39.7	38.5	33.1
Cranberry	7.2	11.2	9.9
Black currant	18.7	25.9	20.3
Wines			
Concord white	7.8	12.1	10.1
Concord red	9.1	13.5	10.1
Bordeaux white	6.0	9.5	7.0
Bordeaux red	4.7	10.9	7.2
Apple	6.0	9.1	6.8
Strawberry	11.0	14.2	11.3
Red currant	9.4	11.6	8.7

[a]Glc of TMS derivative after lead salt precipitation. [b,c]After and before cation exchange.

of this method are the problem of complete precipitation of lead anions and subsequent derivative formation in a heterogeneous mixture and the lack of measure of all acidic components (volatile acidity, CO_2, sulfates) in some cases, *e.g.*, wines.

Phosphoric Acid. One acid component in most juices and all wines bears mention, phosphoric acid. Such a compound has been shown recently (Butts and Rainey, 1971) to readily form the TMS derivative and be eluted on glc, along with a variety of other inorganic anions. Its presence in wine and juice and importance in fermentation process is known (Amerine and Cruess, 1960) but its measurement by glc in fruit products has been reported only recently (Ryan, 1972). As the retention times of this acid as its TMS derivative are very similar to those of succinic, fumaric, malonic, and maleic acids on both SE-30 and OV-17, it can be easily confused with them unless supplementary evidence is available.

As a simple method was available to measure the inorganic phosphate content of juices and wines by glc, a comparison was made between this method and the molybdenum blue method for phosphorus. The latter has been described by Vandercook and Guerrero (1969), the only modification being, in the case of digestion for measurement of the total phosphorus, the sample was subsequently neutralized to pH 8.2. Results for these two methods for apple and grape juices are given in Table IV and are reported as elemental phosphorus.

Table IV. Comparison of Phosphorus Methods, mg of P/100 ml of Juice

	Glc	Molybdenum blue	
	inorganic	Inorganic	Total
Grape juice	9.1	7.9	8.7
	8.1	7.8	8.7
	10.0	8.1	8.5
	9.6	8.6	9.0
	3.7	3.2	3.1
Apple juice	7.7	7.5	
	6.0	5.7	
	5.9	5.6	
	6.6	7.9	
	7.5	6.6	
	6.3	6.2	

The values for P, as measured by glc–TMS, are comparable to and slightly higher than those from the molybdenum blue method. In the case of grape juice the inorganic phosphate represents over 90% of the total phosphorus present, *i.e.*, the P is largely in the PO_4^{3-} form and little is combined in organic forms. Such a relationship was also found with orange juice (Vandercook and Guerrero, 1969). In comparing the merits of the two methods it is to be noted that the glc procedure also gives information about the amounts as well as the kinds of other acids present in the sample.

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