Detection of Adulteration of Raspberry Purees Using Infrared Spectroscopy and Chemometrics

E. K. Kemsley,* J. K. Holland, M. Defernez, and R. H. Wilson

Institute of Food Research, Norwich Laboratory, Norwich Research Park, Colney, Norwich NR4 7UA, United Kingdom

Fourier transform infrared (FTIR) spectroscopy and attenuated total reflection (ATR) sampling have been used to detect adulteration of raspberry purees. A database of 871 spectra of pure and adulterated fruit purees was collected between 1993 and 1994. Partial least-squares (PLS) regression of the spectra onto a dummy variable representing sample type was performed. A 95% confidence interval for the prediction values for pure raspberries was defined: within this region, spectra were accepted as pure raspberry; outside this region, spectra were rejected. Using this criterion, 95% of pure raspberries were accepted as such. Adulteration with apple and plum could be detected at minimum levels of ~20% w/w, with sucrose at ~4% w/w. Spectra of 150 additional samples harvested during 1995 served as further validation samples: comparable classification success rates were obtained. The speed of FTIR spectroscopy makes this technique a rapid method for screening raspberry purees for adulterants.

Keywords: *Infrared; spectroscopy; fruit; purees; adulteration*

INTRODUCTION

The authentication of food materials is of primary importance at all links in the food chain, from raw ingredients to finished products. Many fruit-based products, such as jams, jellies, marmalades, and a variety of other wares, are prepared from fruit purees. These are generally supplied to the manufacturer in frozen form or as sulfited pulps, and consequently their authenticity can be difficult to establish. Fruit purees are often of high value, and the temptation for the supplier to add foreign material, such as sugar syrups or quantities of another cheaper fruit, can be considerable. There is much interest within the food industry in rapid techniques for addressing problems of this nature. Fourier transform infrared (FTIR) spectroscopy is potentially one such technique (Wilson and Goodfellow, 1994). It can be thought of as a molecular "fingerprinting" method: an infrared spectrum contains features arising from vibrations of molecular bonds, and the mid-infrared region (400−4000 cm^{-1}) in particular is highly sensitive to the precise chemical composition of the sample. FTIR has been shown to be useful for a range of identification problems in the food sector (Lai et al., 1994; Briandet et al., 1996), including the classification of purees of different fruit species (Defernez et al., 1995).

In this paper, we present a new method for the detection of adulteration of raspberry purees. Although there are many potential adulterant materials, likely candidates are believed to be sucrose, apple puree, and plum puree. The quantities of adulterant involved may be quite large, in certain cases as much as 90% w/w. Over the past 2 years, we have been conducting a program of research to ascertain the potential of FTIR spectroscopy for tackling this type of problem. We have collected over 1000 spectra of different types of fruit puree. These include pure fruits of different species (raspberry, plum, apple, strawberry, blackberry, cherry, apricot, and black currant), as well as raspberry purees that have been deliberately adulterated with various quantities of sucrose, apple, or plum. The majority of samples have been prepared from fresh or freeze−thawed fruits, harvested by ourselves from sites across the United Kingdom; in addition, some samples were prepared from freeze−thawed whole fruits supplied to us by industrial collaborators. The authenticity of all samples was therefore assured.

A key objective of this research program has been the investigation of suitable data processing methods. Spectra obtained by Fourier transform (FT) methods invariably contain hundreds or even thousands of data values, and chemometric techniques are generally required to fully exploit data of this nature. One such method is partial least squares (PLS) (Martens and Naes, 1989). In its basic regression form, PLS models the relationship between two data sets using a series of linear least-squares fits. It is particularly useful for overcoming some of the problems that are encountered when conventional maximum likelihood methods are applied to large, intercorrelated data sets. Historically, PLS has been used mostly for calibration, for example, to establish relationships between spectroscopic and compositional data. However, we have had good successes in PLS-based discriminant analysis (Kemsley, 1996), in which the experimental data are regressed onto binary "dummy" variables that indicate class membership [regression onto dummy variables is an accepted approach to classification problems (Green, 1978)]. This approach was found to be effective for classifying infrared spectra of olive oils of different types and of different varieties of certain plant species. In the present work, we use PLS regression onto a dummy variable to distinguish spectra of raspberries from those of "non-raspberries" (in which we include the adulterated raspberry samples as well as the other pure fruit samples).

* Author to whom correspondence should be addressed [fax (+44) (0)1603 507723; e-mail kate.kemsley@bbsrc.ac.uk].

S0021-8561(96)00089-1 CCC: $12.00

Table 1. Description of Samples

database					additional samples		
details	fruits harvested	training set samples	tuning set samples	test set samples	details	fruits harvested	samples
raspberry	1993, 1994	1−55	1−53	1−51	raspberry	1995	1−5
blackberry	1993	56−59	54−57	52−55	plum	1995	6−10
plum	1993	60−64	58−62	56−59	raspberries adulterated with 5, 10, 20, 30, 40, 50% w/w plum	1995	11−40
strawberry	1993	65−89	63−87	60−83	mixtures of sucrose, glucose, fructose, and maltose in solution (total sugars content 30−40 g/100 mL)	n/a	41−60
black currant	1993	90−97	88−95	84−90	raspberry	1995	61−96
strawberry	1993	98−135	96−132	91−127	strawberry	1995	97−152
apple	1993	136−158	133−155	128−150			
apricot	1993	159	156	151			
cherry	1993	160					
mixtures of strawberry and apple	1993	161−163	157−159	152−153			
raspberries adulterated with 10, 30, 50, 70 90% w/w apple	1993	164−171	160−167	154−161			
mixtures of strawberry and apple	1993	172−178	168−173	162−168			
mixtures of strawberry and plum	1993	179−181	174−177	169−171			
strawberry	1994	182−227	178−223	172−216			
blackberry	1994	228−232	224−228	217−220			
black currant	1994	233−236	229−231	221−223			
cherry	1994	237−244	232−238	224−230			
plum	1994	245−247	239−241	231−232			
strawberry	1994	248−257	242−250	233−241			
apple	1994	258	251				
apricot	1994	259	252				
raspberries adulterated with 2, 4, 6, 8% w/w sucrose	1994	260−299	253−292	242−280			

EXPERIMENTAL PROCEDURES

Instrumentation. All spectra were collected on a Spectra-Tech (Applied Systems Inc.) Monit-IR FTIR spectrometer system, operating in the mid-infrared (400−4000 cm^{-1}). The instrument was fitted with an air-cooled silicon carbide source, a sealed and desiccated interferometer, and a deuterated triglycine sulfate (DTGS) detector. One of two dedicated sampling stations was equipped with an overhead attenuated total reflection (ATR) accessory, which comprised transfer optics within a desiccated chamber, sealed from the atmosphere by two potassium bromide windows, through which the infrared radiation was directed into a detachable ATR crystal. The zinc selenide crystal was mounted into a plate with a shallow trough for sample containment, and the crystal geometry was 45° parallelogram with mirrored angled faces, and nominally 11 internal reflections.

Samples. In the context of the present work, a single "sample" is defined as a puree of between 6 and 10 fruits, plus quantities of adulterant as appropriate. Purees were prepared from fresh or freeze−thawed whole fruits, by pushing through a fine metal sieve (soft fruits) or blending in a food mixer (Braun Multiquick 300) (tree fruits). In total, 1023 samples were prepared for this work; 871 of these were produced in the period March 1993−March 1995, from fruits harvested in the 1993 and 1994 growing seasons. Spectra of these samples collectively define the "database" from which the PLS regression has been developed. Additional samples (152) were prepared in the period March−September 1995. These include purees of fruits from the 1995 harvest, as well as a range of adulterated samples. Spectra of these additional samples have been used to further demonstrate the robustness and stability of the method. Detailed information on the composition of the database and additional samples is given in Table 1. The database has been randomly subdivided into three subsets, designated "training", "tuning", and "test" sets (to be discussed below).

Spectral Acquisition. All spectral measurements were made at nominal 8 cm^{-1} resolution, with 256 interferograms co-added before Fourier transformation. Single-beam ATR spectra were collected of each sample, transformed to absorbance units using a background spectrum of water, and truncated to 235 data points in the region 899−1802 cm^{-1}. A single-point baseline correction at 1802 cm^{-1} was performed, followed by normalization on the integrated spectral area in the region 899−1802 cm^{-1}. Spectra were stored in this pretreated form for subsequent data processing.

Data Processing. All data processing was carried out using Matlab (The Math Works Inc, Natick, MA) running on a personal computer equipped with a 90 MHz Pentium processor and Microsoft Windows 3.1 operating system. Macros were written in-house for carrying out PLS regression, based on the algorithm for orthogonalized PLS with one dependent variable described in the text by Martens and Naes (1989). The training set was used to obtain a series of regressions of different dimensionalities and the tuning set to identify which of these was the optimum. Using the optimum regression model, an "acceptance" region was estimated, within which the predicted results for spectra of pure raspberries can be expected to occur. Once the data analysis protocol was established, the test set was used as a series of independent samples to confirm that the method had sufficient generalization ability. The 152 additional samples from the period March−September 1995 were used to further validate the PLS regression model.

RESULTS AND DISCUSSION

The baseline-corrected and area-normalized spectra defined as the training set are shown in Figure 1. In our earlier work (Kemsley et al., 1995), we have found that these two pretreatments are useful for removing gross, unwanted instrumental effects from the data. Baseline correction reduces the effect on the spectrum of drift in overall instrument response that can occur between the times of acquisition of the sample and background spectra. The effect of area normalization is somewhat more subtle: it can be regarded as a way of standardizing the path length of the ATR crystal, such that spectra collected on different crystals with, say, disparate numbers of reflections can nevertheless be compared. In the future, it will be important that the spectral database reported here can be transferred to other instruments, hence our choice of this pretreatment.

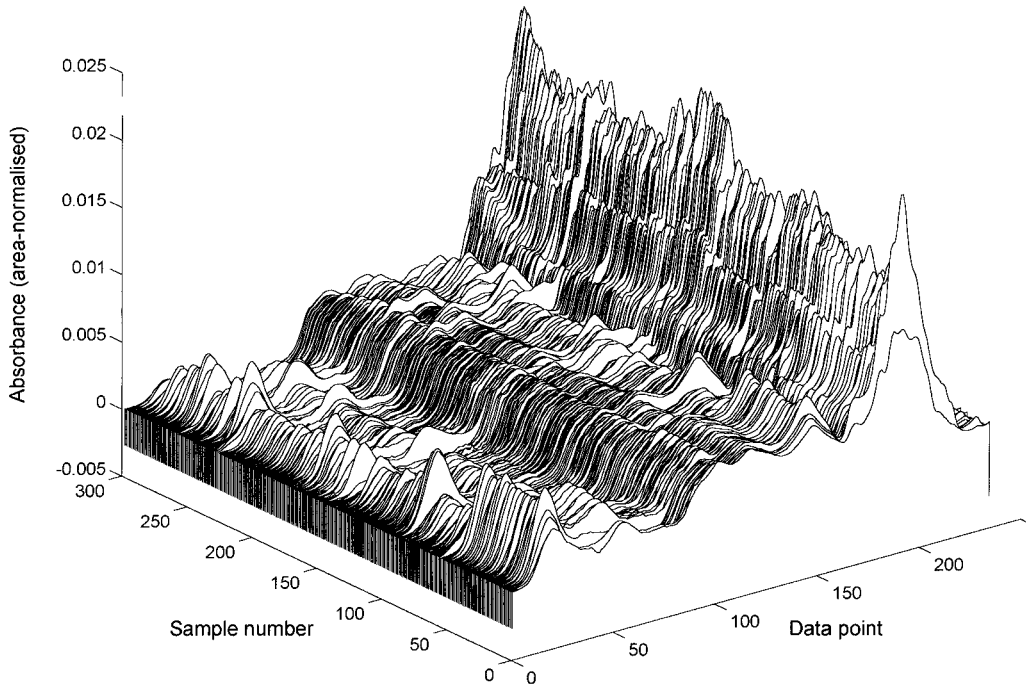

Figure 1. Baseline-corrected, area-normalized spectra of training set samples.

From Figure 1, we obtain an impression of the complexity of the data set and the need for chemometric methods to analyze it. Some preliminary conclusions can nevertheless be drawn from visual inspection. First, there is considerable variability across the entire data set and also within individual fruit species. For example, samples 1−55 are pure raspberries, and even among these spectra there are considerable differences. In addition, certain other fruits yield spectra that are superficially quite similar to those of raspberries: for instance, black currant (samples 90−97 and 233−236 in Figure 1). Some of the spectral features can be attributed to specific chemical constituents. For example, the bands centered on data points ∼20 and ∼60 (corresponding to ∼1725 and ∼1570 cm^{-1}) arise from pectins, and the major features in the region 155−215 data points (1180−950 cm^{-1}) are mainly attributable to carbohydrates. However, a complete spectral assignment is a challenging task, and beyond the scope of the present work. Moreover, the quantities of data involved are too large for much to be accomplished by qualitative analysis, and it is clear that statistical procedures are required to obtain an objective appraisal of these kinds of data.

PLS regression was applied to the training set spectra, using a dummy dependent variable to indicate sample type. The dummy variable is constructed as an ($n \times 1$) vector, where the number of observations n = 299, with entries of 1 for raspberries and 0 for non-raspberries. Let $\mathbf{y}$ be this vector, mean-centered. The spectral data are organized into an ($n \times d$) matrix, where the number of data points d = 235. Let $\mathbf{X}$ be this matrix, again mean-centered. PLS then seeks to relate $\mathbf{X}$ to $\mathbf{y}$ with the model

$$\mathbf{y} = \mathbf{Z}_r\mathbf{p}_r = \mathbf{X}\mathbf{W}_r\mathbf{p}_r \qquad (1)$$

in which $\mathbf{W}_r$ is a ($d \times r$) matrix of PLS loadings and $\mathbf{p}_r$ is an ($r \times 1$) vector of coefficients to be found by multiple linear regression of $\mathbf{y}$ on the ($n \times r$) matrix of PLS scores, $\mathbf{Z}_r$. The subset size r is varied across a range, and the optimum size determined, preferably by applying the regression to a second series of observations. In the present work, these are provided by the tuning set. The vector $\hat{\mathbf{y}}_{tune}$ obtained from applying a regression to the tuning set data is given by

$$\hat{\mathbf{y}}_{tune} = \mathbf{X}_{tune}\mathbf{W}_r\mathbf{p}_r \qquad (2)$$

where $\mathbf{X}_{tune}$ is the matrix of the tuning set data, centered with the mean of the raw training set data. Once the optimum regression model has been established, it is applied to a further series of observations, the test set, from which an estimate of the model's ability to generalize beyond the data with which it was developed can be assessed. The test set can be regarded as truly independent, whereas the tuning set, although not involved in calculating either the PLS loadings or the regression coefficients, is nevertheless involved in decision making concerning regression parameters.

PLS regressions were obtained using r = 5, 10, 15, ..., 50. These were applied to the tuning set data, and a summary statistic calculated: if $\mathbf{y}_{tune}$ represents a dummy variable (again coded according to sample type and centered using the mean of the training set dummy variable), then the quantity ($\mathbf{y}_{tune} - \hat{\mathbf{y}}_{tune}$) represents a vector of residuals, and the standard deviation of its elements can be used as an indicator of the performance of the regression. This statistic was calculated for each regression model and is plotted versus r in Figure 2. The best regressions are obtained for $10 < r < 25$, after which the overfitting regime is entered. The minimum standard deviation of the residuals occurs at r = 15, and this dimensionality was chosen as the optimum.

Figure 3 shows the results of applying this regression model to the training and tuning set data. (For compatibility with the original encoding scheme, the mean of the training set dummy variable has been added to these values; henceforth, it will be assumed that all such "outputs" have been similarly transformed.) In both cases, there is clear differentiation between the raspberries and non-raspberries. Moreover, the general pattern of results for both sets is similar, confirming that the overfitting regime has not been entered.

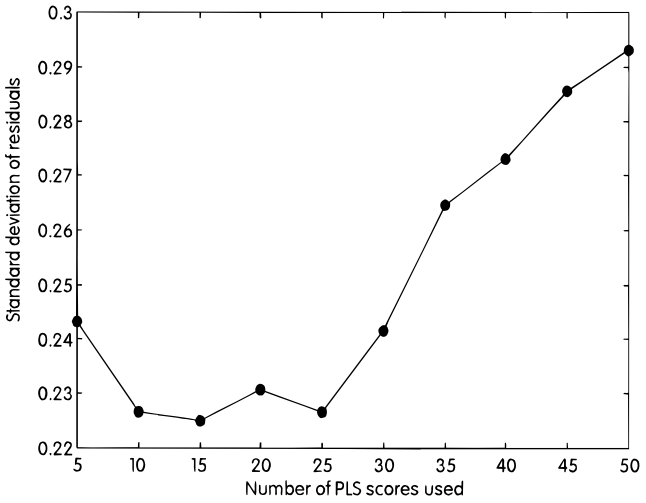

Figure 2. Standard deviation of the tuning set residuals vs number of PLS scores used in the regression.

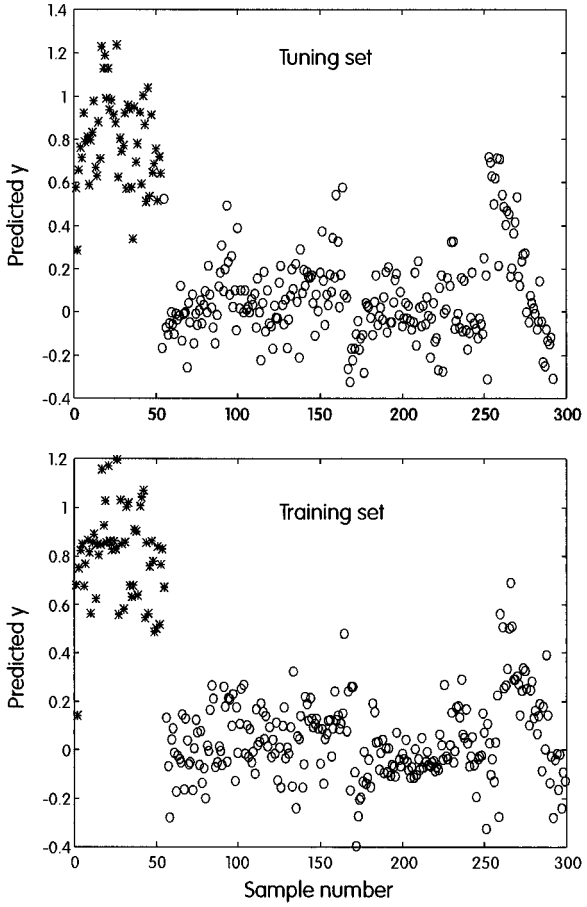

Figure 3. Predicted *y* for the training and tuning sets, using 15 PLS factors in the regression.

A quantitative rule was sought from which decisions on the nature of individual samples could be made. In particular, the outputs for the pure raspberry spectra in both the training and test sets were studied, with the aim of establishing criteria for "acceptance" and "rejection". First, the distributions of the outputs were examined. In linear least-squares regression methods, if a linear model is an appropriate fit for the data, then the residuals should be normally distributed. (In ordinary multiple linear regression, they are ideally normally distributed with a mean of zero; however, PLS is a biased regression method and the residuals will not necessarily have zero mean.) In the present case, in

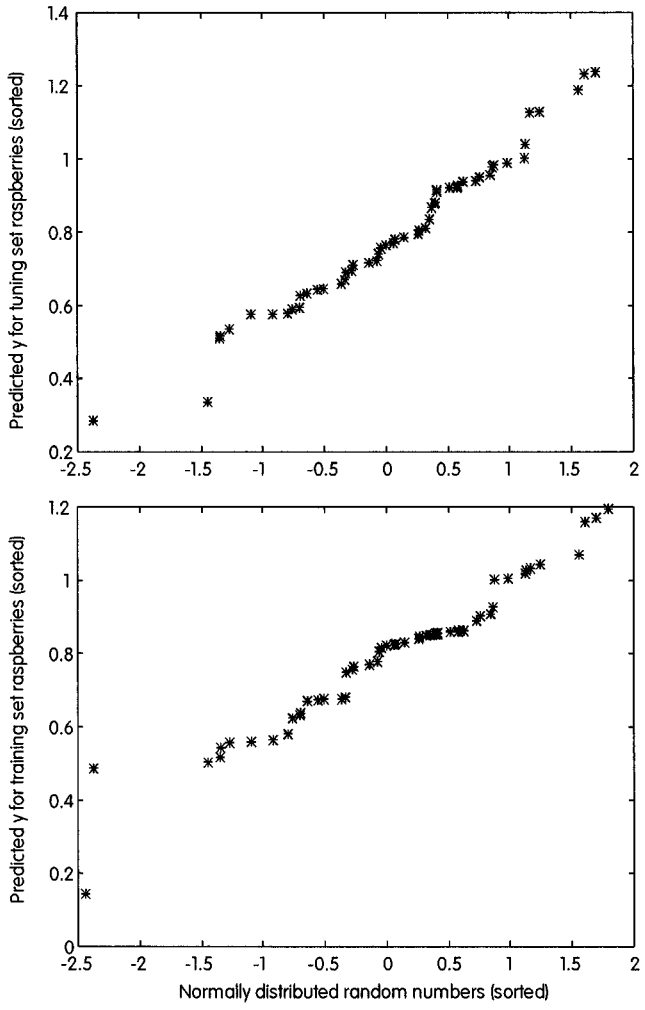

Figure 4. Normal probability plots for the predicted *y* obtained from the training and tuning set raspberry samples.

which the dependent variables take the form of vectors of binary values, the predicted outputs for the raspberries are equal to (1 − residuals); thus, if the linear model is appropriate, the raspberry outputs should also be normally distributed. To test this, probability plots were constructed (Seber, 1984), in which the outputs for the raspberries in each set were sorted into ascending order and plotted against similarly sorted, normally distributed random numbers. We find that these plots are roughly linear (Figure 4), demonstrating that the raspberry outputs can indeed be regarded as normally distributed and hence that linear regression is an appropriate technique. Moreover, the parameters of the distributions describing the outputs from each set are highly similar. For the training set raspberries, the mean output = 0.80 and the standard deviation = 0.19. For the tuning set raspberries, the mean = 0.80 and the standard deviation = 0.21. These parameters are clearly not significantly different from one another, from which we infer that the population to which both sets of outputs belong can be described by the pooled mean $\mu = 0.80$ and pooled standard deviation $\sigma = 0.20$.

Once the parameters of the normal distribution have been established, the construction of confidence intervals is straightforward. The boundaries of the 95% confidence region are $(\mu - 1.96\sigma)$ and $(\mu + 1.96\sigma)$; hence, we propose as the criterion for acceptance of a spectrum as that of pure raspberry, that the output from the PLS regression should fall within the range 0.41−1.19, acknowledging that under these conditions 5% of genu-

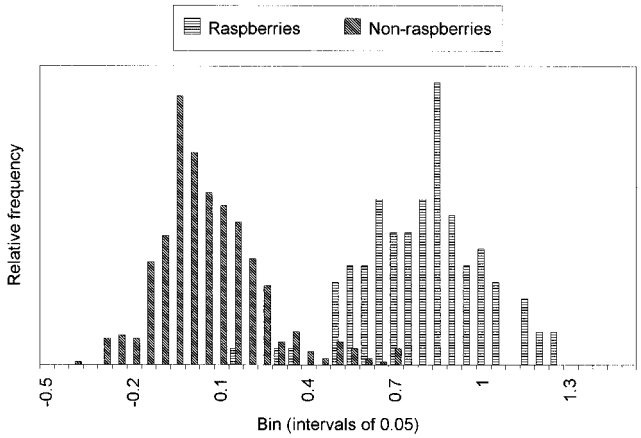

Figure 5. Histograms showing the frequency distributions of the predicted *y* for the combined training and tuning sets.

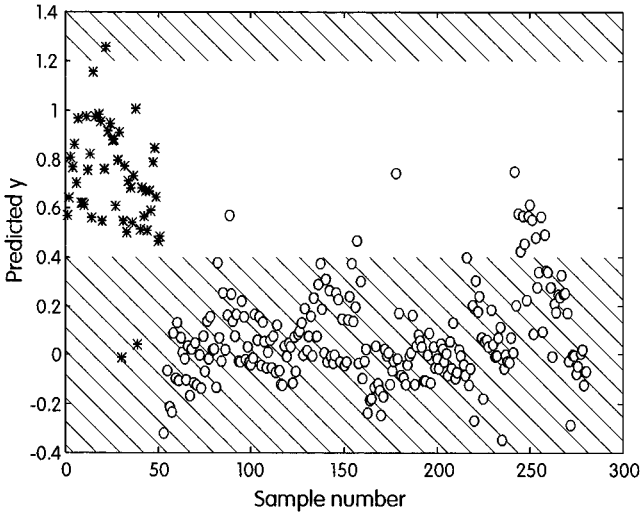

Figure 6. Predicted *y* obtained for the independent test set (95% confidence region corresponds to $0.41 < y < 1.19$) (*, raspberry; ○, non-raspberry).

ine raspberries will be falsely rejected (type I error). The probability of wrongly accepting a non-raspberry (type II error) is much harder to determine. For the current training and tuning sets, we find that the non-raspberry outputs are also approximately normally distributed; Figure 5 shows a histogram of the outputs from the training and tuning sets collectively. If the types of non-raspberries encountered are exclusively those represented in the database, then it is possible to make an estimate of the occurrence rate of type II errors. However, it would be dangerous to assume that this will always be the case; there may exist fruit species or indeed non-fruit adulterants that are chemically even more similar to raspberries, augmenting the chance of making this type of error.

Using a regression based on 15 PLS scores, and accepting as raspberry those spectra with outputs in the range 0.41−1.19, the analysis was applied to the third set from the spectral database, designated the test set. The outputs obtained are illustrated in Figure 6. The rejection zone is shaded. The results are broadly similar to those obtained for the training and tuning sets. A summary of the acceptance rates for each class and for each of the three sets is shown in Table 2. Encouragingly, the acceptance rate for the raspberries averages ∼95%, which is as expected for the confidence interval defined; that is, type I errors are occurring at a rate of ∼5%.

Table 2. Percentage Correct Classifications Obtained for Training, Tuning, and Test Sets

	training set	tuning set	test set	total
raspberries	98.2	92.5	94.1	94.9
non-raspberries	97.5	92.5	93.9	94.6
total	97.9	92.5	94.0	94.8

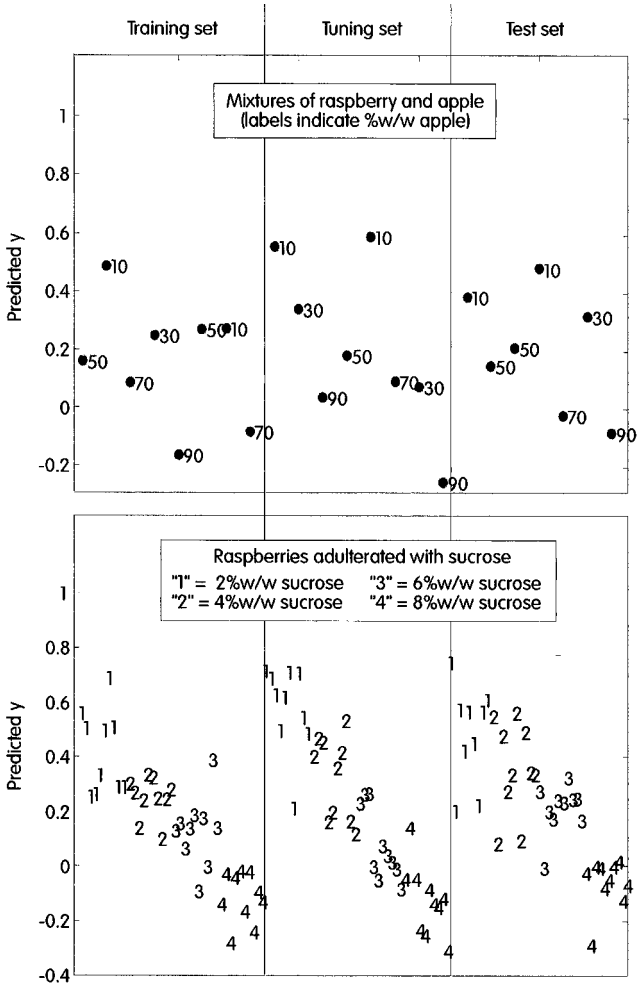

Figure 7. Predicted *y* obtained for the sucrose- and apple-adulterated samples, plotted against sample number for each of the training, tuning, and test sets.

It is worth examining the outputs for certain samples in more detail. Figure 7 collates the results obtained for the apple- and sucrose-adulterated samples in the training, tuning, and test sets. Four of six of the 10% w/w apple-adulterated samples are wrongly accepted as pure raspberry; all samples with higher apple contents are correctly rejected. This suggests that the detection limit for apple is somewhat above 10% w/w; a realistic estimate might be ∼20% w/w. Certainly, at contents of 30% w/w, adulterated samples can be detected. For the sucrose-adulterated samples, the majority of the 2% w/w adulterated samples are wrongly accepted. Most of the 4% w/w samples are correctly rejected, and we suggest that the detection limit for sucrose is around this level. All other samples (6 and 8% w/w) are correctly identified as adulterated. To illustrate the difficulty presented by adulterated samples of this nature, typical spectra of a pure raspberry (training sample 5) and two adulterated samples correctly identified as such (training samples 167 and 275, respectively) are compared in Figure 8. The visible differences between the spectra are minimal and comparable in magnitude to the variations that exist within the series

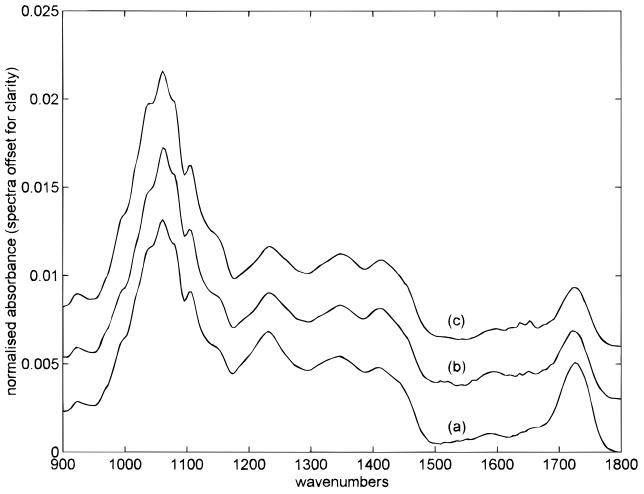

Figure 8. Typical spectra of purees of (a) pure raspberry, (b) raspberry adulterated with 30% w/w apple, and (c) raspberry adulterated with 4% w/w sucrose.

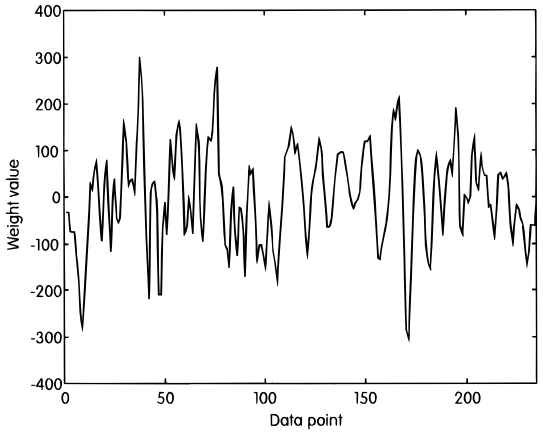

Figure 9. Regression coefficients plotted in "spectral" form.

of pure raspberry spectra. We must therefore conclude that the spectral differences which enable the raspberry and non-raspberry samples to be distinguished are very subtle indeed.

In any regression, it is interesting to examine the coefficients in an attempt to place an interpretation on the nature of the regression model. In the case of PLS regression, we may define a vector of "composite" calibration coefficients **q**, which directly express the relationship between **X** and **y**:

$$\mathbf{y} = \mathbf{Xq}, \qquad \mathbf{q} = \mathbf{W}_r\mathbf{p}_r \tag{3}$$

Thus, **q** represents a single ($d \times 1$) vector onto which mean-centered spectra can be projected to yield the output indicative of sample type. Figure 9 illustrates the coefficients, plotted against data point; the abscissae correspond to those of Figure 1. Unfortunately, it is not possible to place any interpretation on this composite vector, although from the sharpness of the features and the complexity of the structure, we suspect that minor constituents are playing an important role in the analysis.

We will now consider the work that has been carried out since the establishment of the spectral database and data analysis protocol, namely, the application of the methodology to an additional set of raspberry and non-raspberry spectra collected in the period March−September 1995. The results are presented in Figure 10. Of the 41 raspberries, 2 are wrongly classified;

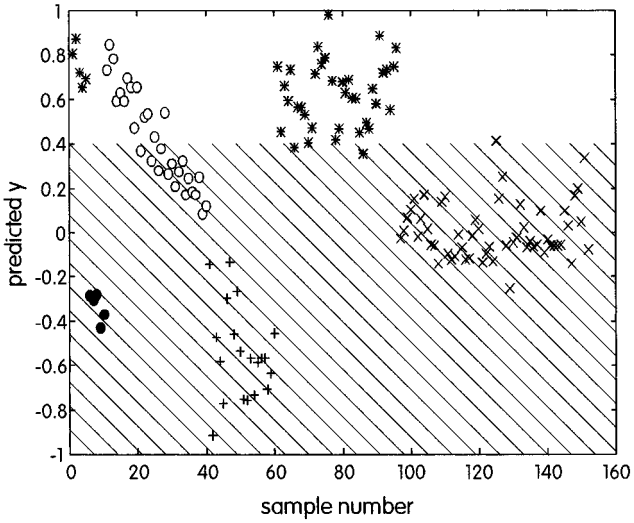

Figure 10. Prediction results for additional samples prepared in the period March−September 1995 (*, raspberry; ●, plum; ○, mixtures of raspberry/plum; +, mixtures of sucrose/glucose/fructose in solution; ×, strawberry).

again, this is approximately 5%, as is to be expected. We conclude that the database of raspberry spectra collected in the previous 2 years is compatible with those from the 1995 harvest, notwithstanding climatic and environmental factors which undoubtedly affect fruit composition, as well as instrumental drift which is likely to be occurring over the time scales involved.

We consider next the mixtures of raspberry and plum. These contained plum in the range 5−50% w/w, and as can be seen, just under half of these samples are wrongly accepted as raspberry. On closer examination of the outputs, it appears that the detection limit for plum is ∼20% w/w. Whether this could be improved upon by performing a separate regression just for plum adulteration is a question we will seek to answer in the future. However, our preferred strategy is a holistic approach, since it is impractical to establish a separate database for each potential adulterant. It is encouraging, in fact, that the present protocol is able to reject mixtures with as little as 20% plum, even though no plum-adulterated samples were present in the database. Similarly, the regression was challenged with a series of spectra of sucrose, glucose, maltose, and fructose mixtures in solution. While these samples neither physically nor chemically resemble fruit purees, their spectra do share many similar features, since carbohydrates exhibit relatively strong absorptions in the infrared and are also major constituents of fruits. Reassuringly, the sugar solutions are all comprehensively rejected by the analysis. Finally, we note that of the 1995 harvest of 56 strawberries, only 1 is wrongly accepted as a raspberry; again, this is consistent with the rejection rate obtained for the comparable database samples.

CONCLUSIONS

In this work we have shown that it is possible to distinguish infrared spectra of raspberry purees from those of a range of non-raspberry materials, including purees of other fruit species, mixtures of various sugars in solution, and raspberry purees adulterated with various proportions of non-raspberry material. The chemometric method chosen for this work was PLS regression onto a dummy variable encoded according to sample type. From the results obtained for the training and tuning sets, we have demonstrated that this linear

method is appropriate. Through the use of the independent test sets, we have shown that the method is able to generalize beyond the spectra with which it was trained and is acceptably stable over time. We believe that the approach taken to encoding the dummy variable (raspberry samples encoded as "1", the wide range of impure or non-raspberries all encoded as "0") means that the regression model works by positively identifying pure raspberry spectra and is relatively insensitive to the precise nature of non-raspberries. We are optimistic that this approach will maximize the chance of a successful outcome when adulterated or non-raspberry material not included in the original database is encountered. There is some evidence to support this premise, in the form of the results obtained when the analysis was presented with spectra of sugar solutions: all were clearly identified as non-raspberry.

Certain potential adulterant materials were studied in more detail: sucrose and apple and plum purees. We estimate that the detection limits for these are ~20% w/w for apple and plum and ~4% w/w for sucrose. These are representative of levels at which adulteration may occur. However, the real power of the present analysis lies in its utility as a rapid screening method: collection of an infrared spectrum takes a few minutes only, and we believe that the regression model described here is likely to be sensitive to a broad range of non-raspberry material.

ACKNOWLEDGMENT

We thank the members of the U.K. Preserves Manufacturers Association for supplying a number of the samples used in this work.

LITERATURE CITED

Briandet, R.; Kemsley, E. K.; Wilson, R. H. Discrimination of arabica and robusta in instant coffee by Fourier transform infrared spectroscopy and chemometrics. *J. Agric. Food Chem.* **1996**, *44*, 170−174.

Defernez, M.; Kemsley, E. K.; Wilson, R. H. The use of infrared spectroscopy and chemometrics for the authentication of fruit purees. *J. Agric. Food Chem.* **1995**, *43*, 109−113.

Green, P. E. *Analyzing Multivariate Data*; Dryden Press: Hinsdale, IL, 1978; Chapter 1 ("The Process of Data Analysis"), pp 10−11.

Kemsley, E. K. Discriminant analysis of high-dimensional data: a comparison of principal components and partial least squares data reduction methods. *Chemom. Intell. Lab. Syst.* **1996**, 33 (1) 47−61.

Kemsley, E. K. Ruault, S.; Wilson, R. H. Discrimination between coffea arabica and canephora variant robusta beans using infrared spectroscopy. *Food Chem.* **1995**, *54* (3), 321−326.

Lai, Y. W.; Kemsley, E. K.; Wilson, R. H. Potential of Fourier transform infrared spectroscopy for the authentication of vegetable oils. *J. Agric. Food Chem.* **1994**, *42*, 1154−1159.

Martens, H.; Naes, T. *Multivariate Calibration*; Wiley: Chichester, U.K., 1989; Chapter 3 ("Methods for Calibration"), pp 116−165.

Seber, G. A. F. *Multivariate Observations*; Wiley: Chichester, U.K., 1984; pp 542−544.

Smith, J. A.; Runtz, L. A. Chemometrics with attenuated total reflectance spectroscopy to detect common adulterants in orange juice concentrate. In *Methods to Detect Adulteration of Fruit Juice Beverages;* Nagy, S., Wade, R. L., Ed.; AgScience: Auburndale, FL, 1995; Vol. 1.

Wilson, R. H.; Goodfellow, B. G. Mid-infrared spectroscopy. In *Spectroscopic Techniques for Food Analysis*; Wilson, R. H., Ed.; VCH: New York, 1994.

Received for review February 8, 1996. Revised manuscript received August 22, 1996. Accepted August 28, 1996.[⊗] The BBSRC, MAFF, and the European Union are acknowledged for financial support.

JF960089L

[⊗] Abstract published in *Advance ACS Abstracts,* November 1, 1996.