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Detection and Quantification of Apple Adulteration in Diluted and Sulfited Strawberry and Raspberry Purées Using Visible and Near-Infrared Spectroscopy

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Adulteration of sulfited strawberry and raspberry purées by apple is a commercial problem. Strawberry ($n = 31$) and raspberry ($n = 30$) purées were prepared from Irish-grown fruit and adulterated at levels of 10−75% w/w using cooking apples. Visible and near-infrared transflectance spectra were recorded using a 0.1 mm sample thickness. Classification and quantification models were developed using raw and scatter-corrected and/or derivatized spectral data. Classification as pure strawberry or raspberry was attempted using soft independent modeling of class analogy. The best models used spectral data in the wavelength ranges 400−1098 nm (strawberry) and 750−1098 nm (raspberry) and produced total correct classification rates of 75% (strawberry) and 95% (raspberry). Quantification of apple content was performed using partial least-squares regression. Lowest predictive errors obtained were 11.3% (raspberry) and 9.0% (strawberry). These results were obtained using spectral data in the wavelength ranges 400−1880 and 1100−1880 nm, respectively. These results suggest minimum detection levels of apple in soft fruit purées of approximately 25 and 20% w/w for raspberry and strawberry, respectively.

KEYWORDS: Strawberry; raspberry; sulfited fruit purée; adulteration; discriminant analysis; quantification; partial least-squares regression

INTRODUCTION

Industrial manufacture of fruit-based foods such as jams, preserves, yogurts, and pie fillings often involves the use of soft fruit purées. Strawberry (*Fragraria ananassa*) and raspberry (*Rubus idaeus*) are two of the most popular such fruits, and global crop production and utilization have been growing to meet food processors' demands. Estimates for global production in 2002 were 3.24×10^6 metric tonnes (strawberry) and 4.14×10^5 metric tonnes (raspberry) (*1*). Chile and the central and eastern European states continue to be the major exporting countries.

Soft fruits are expensive, highly perishable commodities, and their storage and transport have traditionally been in the form of either block-frozen whole fruits or purées. Purées are less expensive than frozen whole fruit and therefore represent the most common method of storage and transport, particularly for processed foods such as jams and pie fillings in which the textural attributes of whole fruits are not of primary importance. Purées are defined as "the edible part of the whole fruit—less the peel, skin, seeds, pips, and like—which has been reduced to a purée by sieving or similar process", although traded purées may sometimes contain seeds (*2*). Because of the loss of most of the original physical structure of the fruit and a considerable reduction of the color intensity on purée manufacture, visual identification of specific fruit types is difficult in this physical form. These losses are all the greater when the fruit is sulfited during purée manufacture in order to increase shelf life, as is the practice in some exporting countries. This is because of the marked reduction in color intensity of purées due to the strong binding of HSO_3^- to the anthocyanin chromophores responsible for their natural color. In such sulfited purées, the opportunities for economic adulteration of strawberry and raspberry purées are even greater than those which present themselves in the untreated product, and such adulteration is believed to be common (*3*). In particular, the adulteration of strawberry and raspberry sulfited purées by the addition of a cheaper, plentiful, and largely colorless fruit such as apple becomes almost impossible to detect sensorially. Global apple production was estimated as 57.1×10^6 metric tonnes in 2002 (*1*).

Detection of a marker compound or compounds that should not be present in a food is one strategy which has been deployed to detect adulteration, for example, detection of phloretin 2′-glucoside (*4*), phloretin 2′-xyloglucoside (*4*), or certain phenolic compounds (*5*), is clear evidence of the presence of apple in a fruit product. Another approach involves the measurement of a range of typical food components, for example, amino acids, sugars, hydroxy acids, carotenoids, lipids, or proteins, and a comparison of the results with established, normal concentration ranges (*6*−*14*). However, this latter approach produces outcomes that are often ambiguous because of the magnitude of natural

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variations in food composition arising from varietal, geographic, seasonal, and maturity differences.

Unsulfited fruit purées retain most of the spectral information arising from naturally occurring anthocyanins, chromophores that have the potential to act as authenticity markers (*15*). For example, the main anthocyanin in strawberry (pelargonidin 3-glucoside) is reported to have a lower λ_{max} than its counterpart (cyanidin 3-sophoroside) in raspberry—512 nm as compared to 523−525 nm (*15*). These compounds are not present in apple, but they are rendered largely colorless as a result of sulfite action, thus diminishing their capacity to contribute to authenticity issues involving soft fruit. Separation techniques normally used to detect these analytes, for example, HPLC or gas chromatography, are time-consuming, expensive, and destructive of the sample. Spectroscopic measurements in the visible and near-infrared regions have been studied with a view to detecting and quantifying food adulteration (*16*). Mid-infrared spectroscopy has also been used with some success (*17*−*20*). The work reported in this paper describes the application of visible and near-infrared spectroscopy to the detection and quantification of apple adulteration of strawberry and raspberry purées to which sodium metabisulfite has been added as a preservative. It represents an extension of previously reported work involving unsulfited strawberry and raspberry purées (*21*).

MATERIALS AND METHODS

Fruit. Samples of strawberries ($n = 31$) and raspberries ($n = 30$) were obtained directly from local growers in Ireland at various times during the 2002 season. Strawberry varieties included were Elsanta ($n = 20$), Cambridge Favourite ($n = 5$), Everest ($n = 1$), Florence ($n = 1$), Symphony ($n = 2$), and Bolero ($n = 1$). Information on the varietal identity of raspberry samples was difficult to obtain, but of those for which this information was available ($n = 14$), the varietal composition was as follows: Glenmoy ($n = 5$), Glenprosen ($n = 1$), Autumn Bliss ($n = 7$), and Glenmagnet ($n = 1$). Nine raspberry samples were from the 2001 season with the remainder harvested in 2002. Whole fruit samples were frozen at −20 °C within 24 h of collection and stored for periods of 6−9 months at this temperature before use. Before being puréed, fruit was thawed at 4 °C for 16 h and the green sepals were removed. Individual fruit samples were puréed in a blender (Braun hand-held liquidizer, 250 W) for 30 s. Immediately thereafter, the purée solids content was measured by refractometry and adjusted to 4% using a freshly made sodium metabisulfite solution containing 1600 ppm of sulfite. Prior to adulteration or spectral collection, purées were stored in covered plastic beakers at 4 °C for no longer than 4 h. Cooking apples labeled as Irish were purchased in local retail outlets and used within 2 days of purchase. Apples were peeled and cored, with only the flesh being puréed. As with the soft fruits, the solids content of apple purées was adjusted to 4% using the aforementioned sulfite solution. Adulterated, sulfited purées ($n = 305$) were produced by adding sulfited apple purée to each sulfited, unadulterated soft fruit purée sample to produce apple concentrations of 10, 20, 30, 50, and 75 wt %.

Spectra. Transflectance spectra (between 400 and 2498 nm at 2 nm intervals) were recorded using a NIRSystems 6500 scanning monochromator (FOSS NIRSystems, Silver Spring, MD) equipped with a sample transport accessory. A camlock transflectance cell fitted with a gold-plated reflector (0.1 mm sample thickness; FOSS U.K. Ltd., P/N 619378) was utilized for sample presentation to the instrument; seeds were manually removed from purée samples prior to closure of the camlock cell. Spectra were collected at ambient temperature (between 18 and 24 °C) in the period from May to July 2003. Two subsamples of each purée were scanned in duplicate with the sample cell being rotated through ∼120° between duplicate scans of each subsample. Mean spectra were used in subsequent data analysis. Spectrophotometer control and preliminary spectral file management was performed using WINISI software (version 1.04a; Infrasoft International, Port Matilda, MD). Exploratory data analysis, calibration development, and validation were performed using WINISI, The Unscrambler (v. 7.6; CAMO A/S, Trondheim, Norway) and Minitab software (release 13.32; www.minitab.com).

Data Analysis. Preliminary data evaluation was performed using principal component analysis (PCA) to detect unusual or outlying spectra. Classification studies were performed using soft independent modeling of class analogy (SIMCA), whereas apple quantification models were developed using partial least-squares (PLS) regression. Each SIMCA and PLS analysis was performed using spectral data in a number of wavelength ranges, that is, 400−750 nm (visible), 750−1100 nm (near near-infrared), and 400−1100, 1100−1880, and 400−1880 nm. The upper wavelength cutoff of 1880 nm was selected due to the high absorbances in purée spectra seen beyond this data point and the concomitant likely nonlinearity of detector response. Calibration models were developed using raw spectral data and after either a scatter-correction pretreatment [standard normal variate (SNV) + detrend (*22*)] or scatter correction plus derivatization procedures (first and second derivatives with a range of gap sizes). The most accurate models involving derivatization used a second-derivative pretreatment step with a 10-datapoint gap size; only those models have been reported in this paper. Class cutoff limits in SIMCA were set at the 5% level; that is, assuming the data are normally distributed, 5% of samples belonging to any given class may be identified as not belonging to it. Classification models (SIMCA) for each soft fruit were developed using 20 of the relevant set of unadulterated purée samples; model evaluation was performed using a spectral file containing all of the fruit purée spectra, that is, all unadulterated and adulterated samples ($n = 366$). The calibration files for strawberry included varieties Elsanta ($n = 8$), Everest ($n = 1$), Symphony ($n = 3$), Cambridge Favourite ($n = 5$), Florence ($n = 1$), and unknown ($n = 2$). Varietal composition of the corresponding raspberry calibration file was Glenmoy ($n = 3$), Autumn Bliss ($n = 4$), and unknown ($n = 13$). Quantitative models for the prediction of apple purée content were developed in WINISI software using modified PLS regression with full leave-one-out cross-validation.

RESULTS AND DISCUSSION

Spectra. Transflectance spectra of representative samples of sulfited apple, strawberry, and raspberry purées are shown in **Figure 1a**. The main features of the spectra are peaks around 1450 and 1940 nm arising from water absorptions. Temperature effects on the spectra of samples containing high contents of water such as these purées can be severe; in this work, these effects were minimized through standardization of the sample preparation and spectral collection rather than through any temperature equilibration step. The similarity between the purée spectra at all wavelengths is marked, as also is the flat peak centered around 2000 nm, which is the result of signal saturation. As mentioned earlier, an upper wavelength cutoff of 1880 nm was selected so as to avoid complications arising from response nonlinearities, even though this has the effect of losing potential information about sugar moieties which occur in the wavelength region 2000−2500 nm. In spectra of unsulfited purées (**Figure 1b**) there is considerable detail in the visible wavelength range, and differences in absorption peak location and size are clearly evident between apple, strawberry, and raspberry fruits (*21*). The addition of sulfite to the purées and the reduction to a solids level of 4% remove spectral detail from the strawberry and raspberry samples in the visible region and reduce absorption peak heights. Given that inclusion of spectral data in the visible wavelength range was a feature of the most accurate models previously reported for the classification and quantification of apple adulteration in unsulfited strawberry and raspberry purées (*21*), it is to be expected that the reduction in the information content in this wavelength range in the case of sulfited purées will have an impact on the success of this analytical approach. The lack of detail above 2000 nm in the spectra of diluted and sulfited purées is also in marked contrast to spectra of untreated purées (*21*).

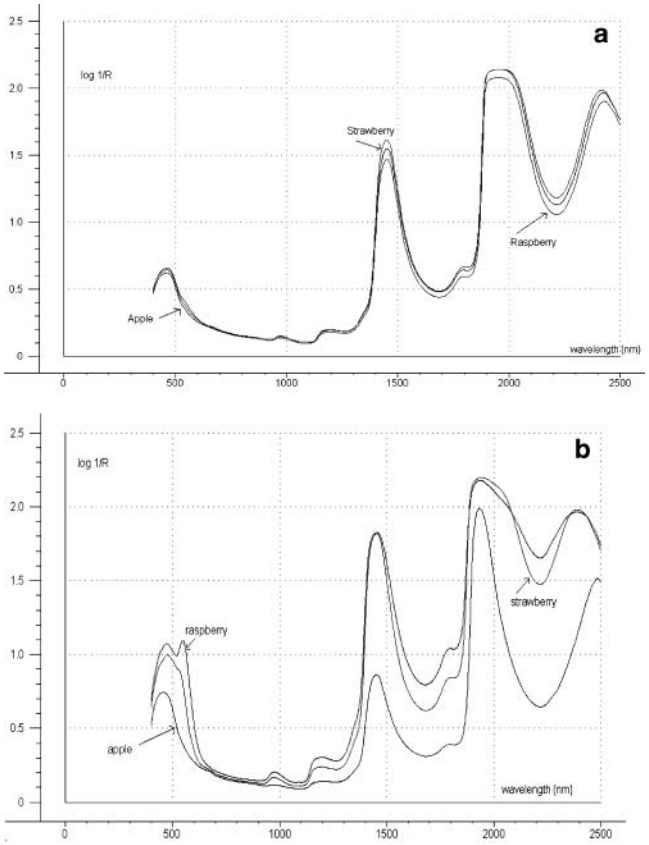

Figure 1. Representative transflectance spectra of (**a**) sulfited apple, raspberry, and strawberry purées (4% solids content) and (**b**) unsulfited apple, raspberry, and strawberry purées (4% solids content) (Reproduced by permission of NIR Publications).

Classification. The accuracy of classification models was assessed on the basis of the number of false positive and false negative results produced by each. A false positive result occurs when a sample is wrongly identified as belonging to a specific class; conversely, a false negative result occurs when a sample that does belong to a class is not classified as such. False positive results may be considered the more serious of the two because they result in failure to detect adulteration or mislabeling. False negatives may be corrected upon subsequent testing using alternative analytical methods. A summary of the classification results obtained by SIMCA for each of the wavelength ranges studied is shown in **Table 1** with the best performing models clearly indicated. These models have generally been selected on the basis that they produce the smallest number of misclassified samples.

In the case of strawberry, it is interesting to note that two models exhibit almost identical performances, that is, those involving the wavelength ranges 400−750 and 400−1098 nm. These models differ only in the distribution of false positive predictions arising from adulterated strawberry and adulterated raspberry samples. In this case, the authors have selected the model using the 400−1098 nm wavelength range as the preferred option on the basis that to rely only on visible wavelength data may weaken the effectiveness of the model because of the relative ease with which purée color characteristics may be manipulated. The importance of the visible wavelength region in these models is interesting, despite the inclusion of sulfite in these purées and its consequent bleaching effect on fruit. No clear pattern concerning, for example, the adulteration level or variety of strawberry samples misclassified by this model was discernible. No unadulterated raspberry samples were misclassified by any of these strawberry models, whereas the level of false negatives was low at 2 or 3 (of 30). Overall, the percentage correct classification for this strawberry model was high at 95.1%.

Table 1. Summary Results of Fruit Purée Classifications [Spectra Collected in Camlock Cell Fitted with Gold-Plated Reflector (0.1 mm Sample Thickness); Most Accurate Models in Boldface Type]

fruit type	wavelength range (nm)	FP[a] overall	FP[a] detail[d]	FN[b]	total misclassifications	PCs[c]
raspberry	**750–1098**	**89**	**28 (S) 61 (AR)**	**2**	**91**	**1**
	400–1880	187	27 (S) + 28 (AS) 132 (AR)	1	188	5
	1100–1880	154	27 (S) + 16 (AS) 111 (AR)	3	157	2
	400–750	193	22 (S) + 43 (AS) 128 (AR)	0	193	4
	400–1098	186	22 (S) + 37 (AS) 127 (AR)	0	186	4
strawberry	750–1098	88	2 (AS) 17 (R) + 69 (AR)	3	91	1
	400–1880	32	12 (AS) 2 (R) + 18 (AR)	3	35	2
	1100–1880	92	17 (AS) 9 (R) + 66 (AR)	3	95	3
	400–750	15	11 (AS) 4 (AR)	2	17	3
	400–1098	**16**	**7 (AS) 9 (AR)**	**2**	**18**	**3**

[a] False positive classifications. [b] False negative classifications. [c] Number of principal components in model. [d] Letters in parentheses describe the false positive samples as follows: S = strawberry; AS = adulterated strawberry; R = raspberry; AR = adulterated raspberry.

In the case of raspberry models, the best performance was achieved using the wavelength range 750−1098 nm (**Table 1**), although the number of misclassified samples is high. The percentage correct classification rate for the best raspberry model was low—75.1%. Interestingly, all of the strawberry samples misclassified by this particular model were unadulterated, whereas other classification models wrongly identified both adulterated and unadulterated strawberry samples. No pattern involving variety or level of adulteration could be detected in the misclassified adulterated raspberry samples.

On the basis of these results, this classification approach does not have the necessary discriminating power to be useful industrially for raspberry purées. However, the strawberry model does possess the necessary classification accuracy to be applied commercially. Interestingly, the best strawberry model reported here is better than that previously reported for similar but unsulfited purées (*21*). In this latter case, although no false negative identifications were made, 36 false positive classifications were reported. No breakdown of the latter according to sample type was presented.

Quantification of Adulterant Level. A total of 15 prediction models was developed for the quantification of apple in unadulterated and adulterated samples of each fruit purée type. These involved five wavelength ranges and the use of three forms of spectral data—raw, treated by standard normal variate and detrending (SNV + detrend), and the latter plus derivation (second derivative, 10-datapoint gap size). The summary results of this work are presented in **Tables 2** and **3** for raspberry and strawberry purées, respectively.

In the case of raspberry, the most accurate model involved SNV + detrend + second-derivative data. This model involved spectral data in the wavelength range 400−1880 nm, used five

Table 2. Summary Results for Quantification of Apple Content in Raspberry Purées [Spectra Collected Using Transflectance Cell (0.1 mm Sample Thickness); Most Accurate Model in Boldface Type]

spectral pretreatment	wavelength range (nm)	loadings[a]	SECV[b]	R^c	a^d
none	750–1098	9	17.5	0.73	1.0
	400–1880	12	11.8	0.89	1.0
	1100–1880	10	12.4	0.87	1.0
	400–750	10	11.5	0.89	1.0
	400–1098	10	12.1	0.88	1.0
SNV + detrend	750–1098	11	15.5	0.79	1.0
	400–1880	11	11.3	0.90	1.0
	1100–1880	11	12.5	0.87	1.0
	400–750	8	11.9	0.88	1.0
	400–1098	8	11.7	0.89	1.0
SNV + detrend[e] + second derivative	750–1098	7	16.5	0.76	1.0
	400–1880	**5**	**11.3**	**0.90**	**1.0**
	1100–1880	7	12.6	0.87	1.0
	400–750	7	11.1	0.90	1.0
	400–1100	7	11.6	0.89	1.0

[a] Number of partial least-squares loadings in regression model. [b] Standard error of cross-validation. [c] Correlation coefficient. [d] Slope of fitted regression line. [e] Standard normal variate and detrending of second-derivative spectra (gap size of 10 data points).

Table 3. Summary Results for Quantification of Apple Content in Strawberry Purées [Spectra Collected Using Transflectance Cell (0.1 mm Sample Thickness); Most Accurate Model in Boldface Type]

spectral pretreatment	wavelength range (nm)	loadings[a]	SECV[b]	R^c	a^d
none	750–1098	13	10.7	0.91	1.0
	400–1880	13	12.1	0.88	1.0
	1100–1880	**11**	**9.0**	**0.94**	**1.0**
	400–750	11	16.2	0.78	1.0
	400–1098	13	10.7	0.91	1.0
SNV + detrend	750–1098	13	9.6	0.93	1.0
	400–1880	13	12.1	0.88	1.0
	1100–1880	13	8.9	0.94	1.0
	400–750	12	14.4	0.83	1.0
	400–1098	13	9.6	0.93	1.0
SNV+ detrend[e] + second derivative	750–1100	12	10.2	0.92	1.0
	400–1880	11	9.1	0.94	1.0
	1100–1880	8	12.1	0.88	1.0
	400–750	5	15.4	0.81	1.0
	400–1100	11	9.1	0.94	1.0

[a] Number of partial least-squares loadings in regression model. [b] Standard error of cross-validation. [c] Correlation coefficient. [d] Slope of fitted regression line. [e] Standard normal variate and detrending of second-derivative spectra (gap size of 10 data points).

PLS loadings, and produced a standard error of cross-validation (SECV) equal to 11.3%. This latter figure suggests that the model is unable to detect apple adulteration at levels below ~23% w/w, that is, 2 × RMSEP. This accuracy figure compares favorably with a detection limit of ~20% w/w for apple in raspberry purées using mid-infrared spectroscopy (*24*). The regression line for this model together with the relevant statistics is shown in **Figure 2**. In total, three models highlighted in **Table 2** produced levels of predictive accuracy that were very similar. For unmodified spectral data, the visible wavelength range (400−750 nm) proved to be most accurate but required 10 loadings. When the spectral data were subject to modification by SNV + detrend only, the best results used the 400−1098 nm wavelength range and required 8 PLS loadings but had a SECV of 11.7%. In this instance, selection of the best model

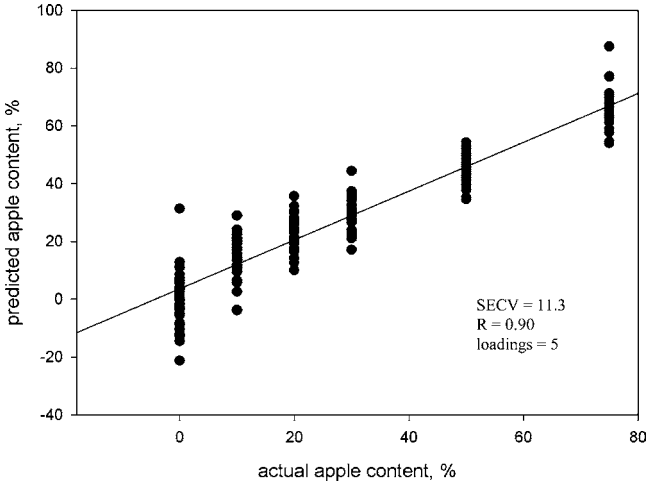

Figure 2. Plot of actual versus predicted apple content in unadulterated and adulterated raspberry purées (400–1880 nm range; SNV + detrend + second derivative using 10-datapoint gap).

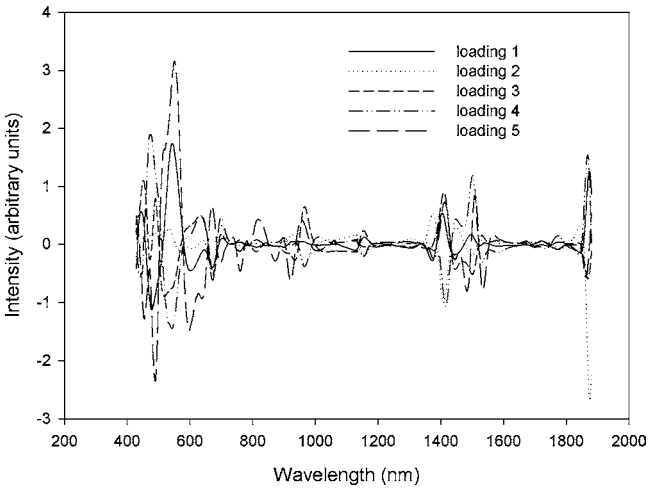

Figure 3. Loadings plot for PLS quantification of apple content in unadulterated and adulterated raspberry purées [400–1880 nm; SNV + detrend + second derivative (10-datapoint gap size)].

involved minimizing the number of PLS loadings required (so as to maximize the chances of model robustness) and avoiding exclusive reliance on visible wavelength data for the reason described above.

Irrespective of the spectral pretreatment used, the wavelength range 750−1098 nm produced the least accurate models. However, inclusion of spectral information from the visible wavelength range was necessary for the most accurate models. This may appear to be surprising given the incorporation of sodium metabisulfite in these purée preparations, but it should be borne in mind that at the levels used, the solutions were not bleached but discolored.

The importance of particular wavelengths in the most accurate model developed for apple quantification is revealed in a plot of the PLS loadings versus wavelength (**Figure 3**). From this graph, it is apparent that considerable information is extracted from the wavelength region 400−750 nm by all five of the loadings, the peak magnitudes of which are significantly greater than those for any other wavelengths in the 400−1880 nm range. There appears to be significant structure in the loadings at a number of regions in the wavelength range, especially around 1400−1600 nm; only the region between 1600 and 1800 nm appears to be relatively flat and therefore limited in significance.

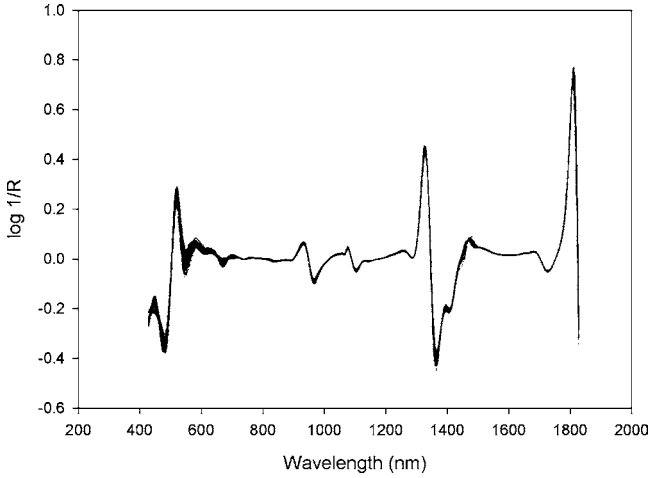

Figure 4. Spectra (400–1880 nm) of raspberry (unadulterated and adulterated) purées after pretreatment [SNV + detrend on second derivative (10-datapoint gap size)].

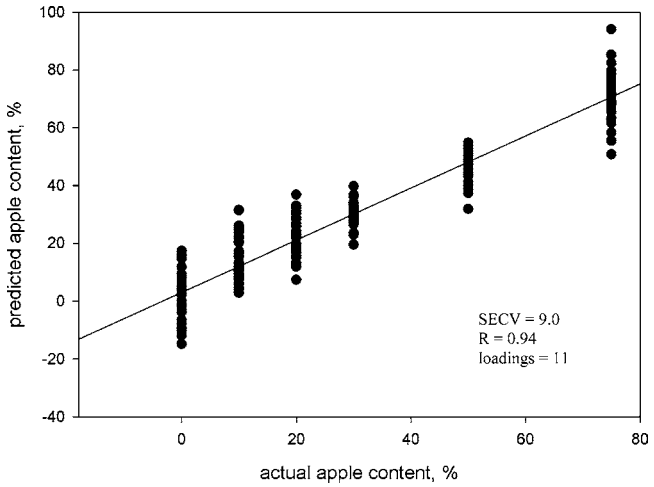

Figure 5. Plot of actual versus predicted apple content in unadulterated and adulterated strawberry purées (raw spectral data; 1100–1880 nm range).

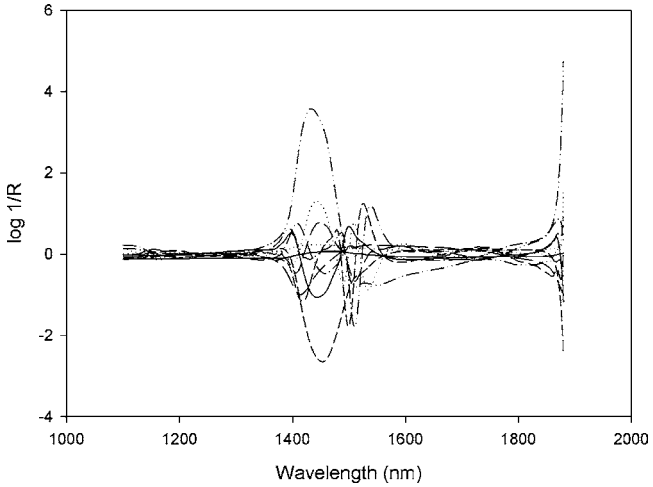

Figure 6. Loadings plot for PLS quantification of apple content in strawberry purées (1100–1880 nm; raw spectral data).

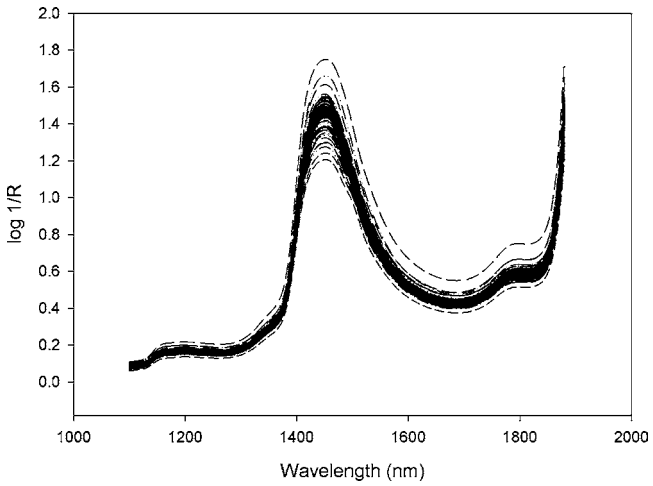

Figure 7. Raw spectra (1100–1880 nm) of strawberry (unadulterated and adulterated) purées.

For comparison, the complete set of spectra from which this model was developed is shown in **Figure 4**; the main features of this graph correspond well with the maxima and minima of the loadings plot (**Figure 3**). Although the involvement of water seems to be indicated by loading peaks and troughs around 1440 and 1800 nm (**Figure 3**), the involvement of other species is reflected by ordered features in the loadings between 1450 and 1600 nm, approximately. These may be related to cellulose, which exhibits a broad peak in this region (*23*). The wavelength range that produced the best qualitative model (750−1098 nm) differed from the quantitative model described here. This may well be explained by the different data compression techniques used in each, that is, PCA (qualification) and PLS (quantification). This difference has been previously observed and reported for unsulfited purées (*21*).

Quantification of apple content in unadulterated and adulterated sulfited strawberry purées was also investigated using the wavelength ranges and spectral data pretreatments mentioned above. A summary of the model performances is shown in **Table 3** with the best model highlighted. In this case, the model of choice predicted apple content with a SECV equal to 9.0%, that is, a limit of detection of 18% w/w (2 × RMSEP). It used raw spectral data in the range 1100−1880 nm (**Figure 5**). Several other models produced prediction statistics that were almost equal to this but on the basis that, all things being equal, simple models are to be preferred to more complex ones, the model using raw spectral data was selected. The main features of the PLS loadings plots (**Figure 6**) occur between 1380 and 1580 nm; this wavelength range was also important in the selected raspberry model and may arise from cellulose as well as water. The involvement of water moieties at around 1800 nm is also apparent (**Figure 6**). Raw spectra of all the strawberry samples in this wavelength range are shown in **Figure 7**.

Conclusions. Adulteration of strawberry and raspberry purées with apple in the presence of sodium metabisulfite is a commercial problem. The discoloration resulting from the inclusion of metabisulfite reduces the information in the visible wavelength range arising from anthocyanin contents, which are characteristic of both soft fruits. This information has been demonstrated to be important in the detection of this type of adulteration in unsulfited purées (*21*). Even in the presence of sulfite, information in the visible wavelength range remains critical for the detection of this form of adulteration for both soft fruits. For routine screening of unknown samples, SIMCA classification was sufficiently accurate to be commercially useful in the case of strawberry (95.1% correct classification). This classification method produced very low correct classification rates for raspberry and is therefore not recommended for industrial use. With regard to apple quantification in purées, PLS regression models produced SECV of 9.0% w/w (straw-

berry) and 11.3% w/w (raspberry). Given the range of adulterant utilized (0−75%), these accuracy levels result in range/error ratios of 8.3 and 6.6, respectively. These standard error levels suggest minimum detectable limits of about 20 and 25% w/w for apple adulteration in strawberry and raspberry purées, respectively. Given that adulteration of raspberry purées by incorporation of ∼20% w/w apple has previously been reported as representative of levels at which adulteration may occur (*24*), the minimum detection limits reported in the present study are appropriate for industrial use.

Both visible and near-infrared spectroscopy have previously been shown to quantify apple content in adulterated unsulfited strawberry and raspberry purées with prediction errors of 5.5 and 3.4% w/w, respectively (*21*). One way of improving the quantification of apple in sulfited purées reported in this work may be to incorporate acetaldehyde in the purée dilution solution, thereby trapping sulfite and restoring the spectral information arising from the soft fruit chromophores. This procedure is based on the stronger binding of acetaldehyde to anthocyanins than to sulfite and is already applied to the measurement of phenolic compounds in wine (*25*). Its application would allow the application of predictive models, either new or based on the previous work on unsulfited purées, which may be expected to possess predictive accuracies better than those developed in this work.

ACKNOWLEDGMENT

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