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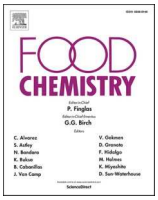

Screening method for the rapid detection of diethylene glycol in beer based on chemometrics and portable near-infrared spectroscopy

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ABSTRACT

A recent case of contamination of some batches of a Brazilian beer brand with diethylene glycol (DEG) had great repercussion, resulting in at least seven deaths. In this article, a direct method was developed for the rapid detection of DEG in beer samples based on portable near-infrared spectroscopy combined with partial least squares discriminant analysis (PLS-DA). The discriminant model was built with 100 uncontaminated beer samples and 100 samples containing DEG in a concentration range between 10 and 1000 mg L^{-1}, totalizing 200 samples of different brands and styles. The method was validated by estimating figures of merit, such as false positive and false negative rates, sensitivity, specificity, accuracy, accordance, and concordance. The decision limit (CCα) of the method was 52 mg L^{-1} and the detection capability (CCβ) was 106 mg L^{-1}. This method does not consume reagents/solvents and can be suitable for the beer industry quality control or forensic investigations.

1. Introduction

The principles of food safety ensure access to food and beverage of adequate quality and quantity for healthy eating, besides being a fundamental and universal human right. These principles have become an increasingly important topic nowadays (Prosekov & Ivanova, 2018). As for other industrialized food products, the quality control of a complex beverage such as beer is also imperative (Sileoni, Marconi, & Perreti, 2015; Fox, 2020; Fulgêncio, Araújo, Pereira, Botelho, & Sena, 2019). The development of new analytical methods jointly with the rapid evaluation of their results for the quality control of the final product is an essential tool for improving brewing techniques. Despite all safety protocols adopted in the brewing production, problems caused by contamination may occur due to several reasons, such as accidental failure, lack of acceptable conditions during production or distribution, negligence, malpractices, or even criminal actions.

At the end of 2019, some unusual cases of acute renal failure with neurological alteration were reported in the State of Minas Gerais, Brazil. In January 2020, a local brewery was implicated in the food poisoning of dozens of people related to these previous occurrences, resulting in at least seven deaths. This case had great repercussion in the Brazilian media, and the brewery production was interrupted. The results of the investigation have elucidated the causes of this tragedy. DEG was employed as antifreeze agent, and a hole in the antifreeze piping of the fermenter tank was detected, through which this substance inadvertently leaked out and contaminated the beer (Goulart, Bordoni, Nascentes, & Costa, 2020; Sanchez, Oliveira, Laranjeira, & Caetano, 2020; Lima, Braga, Ventura, & Goulart, 2021).

Diethylene glycol is a clear, practically odorless, colorless, and viscous liquid, with a sweetish taste and soluble in water and ethanol. Its chemical formula is $(HOCH_2CH_2)_2O$. DEG has numerous industrial uses, mainly as antifreeze agent, chemical intermediary and solvent, but also as heat transfer fluid, brake fluid, cement processing, and lubricant. DEG has a low toxicity for itself, the major problem being its biotransformation in the liver leading to highly toxic metabolites, such as 2-hydroxyethoxyacetic (2-HEAA) and diglycolic acid (DGA) (and possible other yet unknown metabolites). These metabolites may persist in the body for a long time and cause acute toxic syndrome, resulting in renal failure due to cortical tubular degeneration and proximal tubular necrosis. The minimum dose for DEG toxicity in humans has not been clearly established, but the range of doses reported as lethal for DEG has varied from 0.5 to 5 g kg^{-1} (Schep, Slaughter, Temple, & Beasley, 2009; Landry,

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Martin, & McMartin, 2011). Unlike pharmaceutical products and excipients, there is no maximum allowed limit for DEG in food and beverage (Caldeira, Madureira, Maia, Muller, & Fernandes, 2021).

DEG poisoning has been related to incidents involving matrices such as pharmaceuticals, healthcare products and alcoholic beverages. Several analytical methods have been reported in the literature for determining DEG in these types of matrices, mainly based on chromatographic techniques. The most used technique has been gas chromatography with flame ionization detection (GC-FID). GC-FID has been employed to determine DEG in pharmaceutical products (Baffi, Elneser, Baffi, & Melin, 2000), toothpaste (Holloway, Maheswaran, Leeks, Bradby, & Wahab, 2010), food items such as soft drinks, juice, infant formula, cereal, flour and snacks (Rahim et al., 2011), and wine (Lawrence, Chadha, Lau, & Weber, 1986). This last method has been developed because of a real case in which DEG has been detected at levels exceeding 100 mg L^{-1} in wines; the addition of DEG to wine would be made aiming to improve its sweet taste. Ultra-performance liquid chromatography–time of flight mass spectrometry (UHPLC-MS) has been used to determine DEG in toothpaste (Hernández, Ibáñez, & Sancho, 2008) and GC–MS to determine the same analyte in human plasma (Maurer, Peters, Paul, & Kraemer, 2001). GC–MS has also been very recently utilized to detect and quantify DEG in beer by a Brazilian research group, reverberating the contamination occurrence in Minas Gerais (Caldeira et al., 2021).

Despite their advantages, chromatographic methods present some drawbacks, requiring costly, laborious and time-consuming analysis, consuming organic solvents, generating chemical waste, and demanding previous sample preparation steps, such as derivatization, clean up and preconcentration. On the other hand, the emergence of green chemistry has brought up concerns about the development of new more environmentally friendly analytical methods. Vibrational spectroscopic techniques have lower sensitivity in comparison with chromatography, but represent a green, simple and more rapid alternative to develop screening methods. The concomitant use of chemometrics is almost mandatory. Thus, vibrational techniques have been combined with chemometric models, mainly partial least squares (PLS), to detect and quantify DEG in several matrices. DEG has been determined in glycerin with Raman spectroscopy (Gryniewicz-Ruzicka et al., 2011), in glycerin-based cough syrup with mid and near infrared spectroscopies (Ahmed, McLeod, Nézivar, & Giuliani, 2010), and in toothpaste and gel dentifrice with attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy (López-Sánchez, Domínguez-Vidal, Ayora-Cañada, & Molina-Díaz, 2008).

Portability is an important trend in spectroscopy, allowing simple, robust and real-time analysis (Crocombe, 2018). Portable near infrared (NIR) spectrometers have sensitivity and performance similar to that of benchtop equipments, showing other advantages such as small size, lower energy consumption and absence of moving parts in their optical structure. These characteristics improve the usefulness of NIR spectroscopy as an alternative to chromatographic techniques for the quality control of food and beverage. Particularly for brewery production, handheld NIR spectrometers can be viewed as sensors useful to establishing more control of the whole process, reducing production costs, and improving the confidence of the final product. In addition, other parameters can be monitored in the scope of process analytical technology (PAT) (Sileoni et al., 2015; Fox, 2020). It can be used for instance to determine alcohol content (Fulgêncio, Resende, Teixeira, Botelho, & Sena, 2022) and color (Fulgêncio et al., 2019) of beers. Other liquid samples have also been checked for authenticity and quality control with portable devices, such as fresh juice (Chen et al., 2021), palm (Teye, Elliot, Sam-Amoah, & Mingle, 2019) and coriander oils (Kaufmann, Sampaio, García-Martín, & Barbin, 2022). Considering the points previously discussed, the scientific question approached in this article is to evaluate if a handheld NIR device can be used to detect DEG contamination in beer.

Thus, the aim of this study was to combine portable NIR spectroscopy and PLS discriminant analysis (PLS-DA) to develop and validate a rapid and simple screening method for the direct detection of diethylene glycol in beer. Aiming to obtain a robust model, beers from different styles, alcohol contents, brands, and breweries were used to build the multivariate model. The proposed method was validated incorporating multivariate aspects through the estimate of proper qualitative figures of merit (FOM) (Botelho, Reis, Oliveira, & Sena, 2015; The Commission of the European Communities, 2012; Gondim, Junqueira, Souza, Pilar Callao, & Ruisánchez, 2017; Isabel López, Pilar Callao, & Ruisánchez, 2015). FOM related to concentration were also estimated by fitting a probability of detection (POD) curve with the PLS-DA outputs (Gondim et al., 2017; Isabel López et al., 2015).

2. Materials and methods

2.1. Instruments and software

NIR spectra were collected using a portable MicroNIR® 1700 spectrophotometer from Viavi Solution (Milpitas, CA, USA), with a wavelength working range between 900 and 1700 nm. One of the main components of this portable equipment is the linear variable filter (LVF), a part responsible for passing frequencies within the working range and rejecting or attenuating frequencies outside this range. Since the center wavelength (CWL) is chosen as a function of coating thickness, the peak wavelength transmitted through the filter will vary linearly in the wedge direction. Therefore, specifically for the MicroNIR®, wavelengths are selected by adjusting the filter to the appropriate linear position. Thus, the LVF is arranged above the InGaAs array detector. The detector is a variable-band semiconductor with excellent optical properties. The diffusely reflected radiation was collected and sent to a computer. Data were processed using MATLAB® software, version 7.13 (MathWorks, Natick, USA), coupled with the PLS Toolbox, version 6.5 (Eigenvector Technologies, Manson, USA).

2.2. Beer samples

One hundred beer samples were purchased mainly in the local commerce of the Metropolitan Region of Belo Horizonte, Minas Gerais, Brazil, and stored at room temperature until the analysis. These beer samples were purchased from a great variety of brands, breweries, alcohol contents and types, including lager, pale ale, red ale, Vienna lager, Pilsen, IPA, weissbier, strong pale ale, porter, and stout. All samples were previously checked for the absence of DEG using a chromatographic methodology described in the next section.

2.3. GC–MS method for assuring the absence of DEG in non-contaminated beers

A chromatographic method was developed for checking the absence of DEG in non-contaminated beers. Beer samples were sonicated for 5 min, diluted ten times with methanol (HPLC grade), and an aliquot of 100 μL was transferred to a 2 mL Eppendorf. Then, the sample was centrifuged for 5 min at 3000 rpm, and transferred to 2 mL vials, before the injection in the chromatograph. A Shimadzu GCMS-QP2010 Plus system (Kyoto, Japan) was utilized jointly with an Innowax column (60 m × 0.25 100 μm × 0.25 mm). The injection volume was 1.0 μL in the splitless mode, the temperatures of the injection port and the detector were set at 250 °C, and helium was used as carrier gas in a flow of 1.0 mL min^{-1}. The ions of m/z 43 and 45, characteristics of DEG, were monitored. Oven temperature was kept at 100 °C for 1 min, increased to 200 °C at a rate of 10 °C/min, kept at 200 °C for 10 min, increased to 240 °C at a rate of 10 °C/min, and kept at 240 °C for 5 min.

2.4. Sample preparation and spectra acquisition

All the reagents were of analytical grade and used as received. A

volume of 10 mL of each beer sample was degassed using an ultrasonic bath (Equilab ULTRAsonik 28H, Madrid, Spain) for 5 min. One drop of 1-octanol was added to each beer sample to avoid excessive foaming. This step is not feasible for direct application in the industrial brewing process, but it can be adapted for an at-line monitoring. One hundred original beer samples were selected in random order and spiked with DEG in a concentration range from 10 to 1000 mg L^{-1}, with an increment of 20 mg L^{-1} between samples. A volume of 2.0 mL of each sample contaminated with DEG was transferred to a 2 mL Eppendorf tube. For non-contaminated samples, 2.0 mL of each original beer sample was placed in a 2 mL Eppendorf tube without any additional preparation. All the Eppendorf tubes were placed in an acrylonitrile–butadienestyrene support especially designed and constructed for this application with a 3D printer (Creality Ender 3, Shenzhen, China), which helped to collect the spectra of the two hundred samples with the portable MicroNIR® spectrophotometer. Spectra were recorded in the diffuse reflectance mode, with twenty scans each. The wavelength range was from 908 to 1676 nm with a resolution of 6.25 nm. During all these measurements, the temperature of the laboratory was controlled at 25 ± 1 °C. The NIR spectrum of a sample of pure DEG was also recorded.

2.5. Qualitative analytical validation

The analytical validation of the developed model was assessed by estimating appropriate qualitative FOM, such as false-negative rate (FNR), false-positive rate (FPR), sensitivity rate (STR), specificity rate (SPR), accuracy, accordance (ACC), concordance (CON), detection capability (CCβ), and decision limit (CCα). Accuracy is a FOM that expresses globally the rate of incorrect predictions regardless they are positive or negative. ACC and CON express the precision of qualitative methods, being equivalent to quantitative repeatability and intermediate precision, respectively. These FOM have been initially proposed for univariate methods (Langton, Chevennement, Nagelkerke, & Lombard, 2002) and can be directly extended to multivariate methods (Botelho, et al., 2015). ACC was estimated using ten replicates at five different levels (50, 80, 100, 400, and 1000 mg L^{-1}) analyzed in the same batch under repeatability conditions (the same day and the same analyst). CON was estimated using the same protocol employed for ACC, repeated at two different days by two different analysts. CCα and CCβ were estimated from POD curves, which harmonize the statistical concepts and parameters between quantitative and qualitative method validation. This type of curve presents graphically the relation between the probability of positive results obtained from qualitative methods, *i.e.* the outcomes of classification models, and the analyte concentration. The probability values are calculated from independent measurements at each level of adulteration (Isabel López et al., 2015; Gondim et al., 2017). For estimating CCα and CCβ, ten fortified sample replicates were prepared at ten different concentration levels (blank, 10 mg L^{-1}, 20 mg L^{-1}, 40 mg L^{-1}, 60 mg L^{-1}, 80 mg L^{-1}, 100 mg L^{-1}, 400 mg L^{-1}, 700 mg L^{-1} and 1000 mg L^{-1}).

3. Results and discussion

3.1. PLS-DA model building

Beer is a complex matrix. Its main ingredients include water, which is a critical component in the brewing process, as well as hops and yeast, besides other additives such as fruits, herbs, and other plants. Fig. 1 shows the preprocessed NIR spectra, in the wavelength range between 908 and 1676 nm, of the 200 analyzed beer samples, 100 contaminated with DEG and 100 non-contaminated. The absence of DEG in the non-contaminated beer samples was verified by GC–MS (section 2.3). By observing these spectra, it is possible to highlight the two most intense absorption bands, one centered around 1200 nm, and other one centered around 1400 nm. The first spectral band, in the region between 1100 and 1250 nm, may be assigned to the second overtone stretching of CH, CH_2, and CH_3 bonds; the second band might be related to the first overtone of the OH stretching (Metrohm NIRSystems, 2014). More specifically, the first mentioned band can be assigned to C—H stretching overtone of ethanol, while the spectral band at higher wavelengths can be related to O—H bonds of ethanol and water (Fulgêncio et al., 2019). Spectral profiles of all the samples are similar, thus preventing the possibility of discrimination between contaminated and non-contaminated beers by visual inspection. Considering the limited selectivity of NIR spectroscopy, the concomitant use of chemometrics to extract information is imperative. Thus, a multivariate supervised classification PLS-DA model was built.

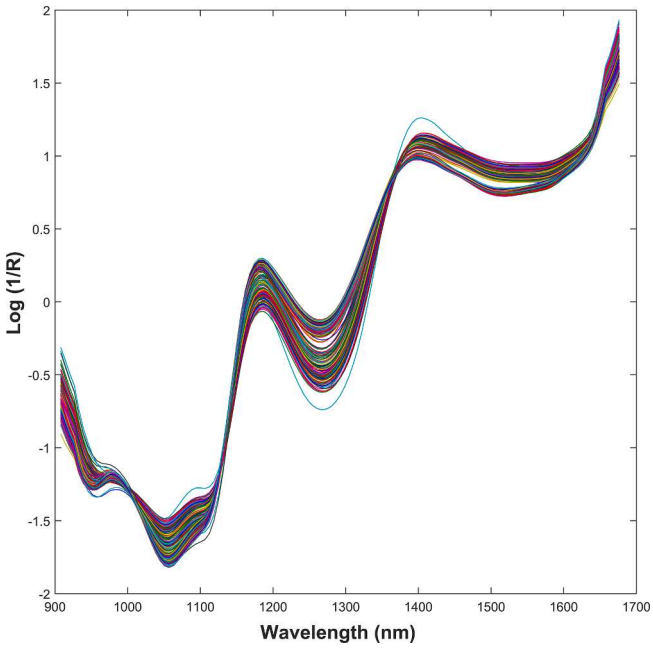

Fig. 1. NIR spectra preprocessed by Savitzky-Golay smoothing and standard normal variate (SNV) of all the 200 analyzed beer samples.

For building the PLS-DA model, samples were split in training and test sets, in the ratio of approximately two thirds:one third. Kennard-Stone algorithm was used for selecting training samples from each of the two classes (Kennard & Stone, 1969). Thus, 134 training samples were selected, whereas the remaining 66 samples were used to construct the test set. Aiming to eliminate drifts typical of diffuse reflectance infrared spectra and caused by multiplicative light scattering, data was sequentially preprocessed by Savitzky-Golay smoothing (7 points and second order polynomial fit), standard normal variate (SNV), and mean centering (Rinnan, van den Berg, & Engelsen, 2009).

Non-contaminated samples were trained with y values equal to 1.0, while samples contaminated with DEG were associated to 0.0. Random subsets cross-validation (6 splits and 20 iterations) was applied to select the number of latent variables (LV), according to the smallest cross-validation classification error (CVCE). The best PLS-DA model was selected with 6 LV after optimization by outlier detection, accounting for 99.63% of the spectral variance (X block) and 71.41% of the dummy variables variance (Y block). In this study, outlier detection was performed based on leverage and Q (spectral) residues, both at 95% confidence level. The number of outliers cannot exceed the recommended limit of 2/9 (22.2 %) for both training and test sets (MAPA, 2014; Thompson, Ellison, & Wood, 2002). Twelve out of the original two hundred samples were removed as outliers, seven from the training set (5.2 %) and five from the validation set (7.6 %). After the removal of outliers, the model was rebuilt. The results for the optimization of this PLS-DA model, including STR and SPR for cross-validation, in addition to CVCE, are shown in Table 1. The Bayesian threshold was estimated at 0.51 and the plot of individual y predicted values is shown in Fig. 2.

Table 1
Results for the optimization by outlier detection of the PLS-DA model built for the detection of diethylene glycol in beer.

		Number of training samples	Number of test samples	Number of latent numbers	Sensitivity (CV)[a]	Specificity (CV)[a]	Cross-validation classification error (CVCE)
Before	DEG-Free	67	33	6	0.851	0.834	0.158
	DEG	67	33				
After	DEG-Free	62	29	6	0.956	0.927	0.059
	DEG	65	32				

[a] Cross-validation.

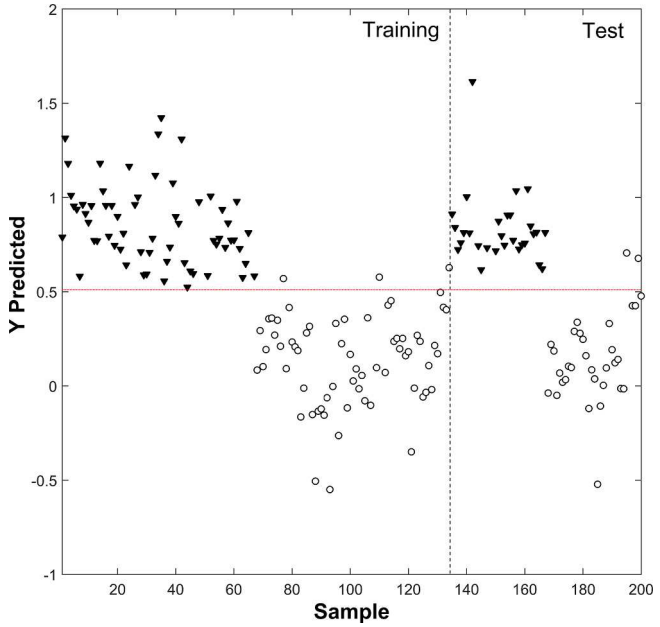

Fig. 2. Y predicted values for the PLS-DA model built to discriminate DEG contaminated and non-contaminated beer samples. Horizontal dashed line indicates the threshold and vertical dashed line indicates the separation between training and test samples. Full down triangles represent non-contaminated samples and empty circles represent samples contaminated with DEG.

3.2. Spectral interpretation of the PLS-DA model

Qualitative or quantitative chemometric models should be corroborated through the spectral interpretation of their informative vectors. The estimated vectors of regression coefficients and variable importance in projection (VIP) scores for the developed PLS-DA model are shown in Fig. 3. VIP scores (Fig. 3a) indicate the variables that most contribute to the model in absolute values (Chong & Jun 2005). Usually, variables with VIP scores higher than 1.0 are considered the most significant for the model. Since these parameters present absolute values, they should be evaluated as contributing to the model as whole and cannot be directly associated with one class. Thus, the spectral interpretation should be complemented by inspecting the regression vectors (Fig. 3b), because they can indicate to which class each variable is more related. Variables with the most positive regression coefficients contribute to the characterization of the class 1 (in this study, non-contaminated beer samples), while variables with the most negative regression coefficients characterize class 0 (beer samples contaminated with DEG). The most discriminant variables do not necessarily coincide with the most intense absorptions. Aiming to complement this spectral interpretation, the NIR spectrum of a pure DEG sample is shown in Fig. S1 (Supplementary Material), jointly with the loadings plot of LV2 in Fig. S2. By observing spectral features, LV2 was assigned to the DEG contribution to the PLS-DA model.

By observing Fig. 3, it was possible to identify the NIR spectral regions that most contribute to the detection of DEG in beer. There are three negative regions of regression coefficients, which are related to the presence of DEG. The most intense and sharp peak, at 945 nm, is related to the third overtone of CH_2 vibrations (Metrohm NIRSystems, 2014) and may be associated with the chemical structure of DEG. The large band of regression coefficients centered at 1484 nm corresponds to an intense and broad absorption band of the NIR spectrum of DEG (Yin, Zhang, Li, & Jin, 2014; Li, Arzhantsev, Kauffman, & Spencer, 2011) and can be assigned to the first overtone of its OH bonds (Ahmed et al., 2010). Finally, the band centered at 1150 nm, which also presents the highest VIP scores, can be assigned to the second overtone of CH_2 bonds and corresponds to a band of lower absorption in the NIR spectrum of DEG (Yin et al., 2014; Li et al., 2011). All these spectral assignments can also be corroborated by observing Figs. S1 and S2. By contrast, the most positive regions of regression coefficients, which also correspond to some of the highest VIP scores, are centered at 1013 and 1410 nm. They can be assigned to the second and first overtones of OH stretching (Metrohm NIRSystems, 2014), which can be attributed to constituents of beer, such as ethanol and water. These signals are direct interferences for the detection of DEG.

3.3. Analytical validation of the PLS-DA model

The analytical validation of the developed method was performed through the estimate of proper FOM. The values for seven different qualitative FOM are shown in Table 2. FNR, FPR, STR, SPR and accuracy were estimated for both training and test sets, while FOM related to precision, ACC and CON, were estimated only for the test set. PLS-DA model did not provide any false negative prediction, leading to STR of 100% for both training and test sets. This means that DEG was correctly not detected in all the non-contaminated samples. On the other hand, three false positive predictions for the training set and two false positives for the test set were observed. These results in SPR of 95.5% and 93.9% for training and test sets, respectively. All these false positives corresponded to samples contaminated with the lowest concentration levels of DEG, thus pointing to a limitation in the sensitivity of the method, which might be related to the limits of the analytical technique, NIR spectroscopy (Pasquini, 2018). Considering the observed limitation of the method to detect the lowest DEG concentrations, FOM related to precision were estimated at five levels above 50 mg L^{-1} (50, 80, 100, 400, and 1000 mg L^{-1}). ACC corresponds to qualitative intra-run precision, while CON is related to inter-run precision. For DEG concentration at 50 mg L^{-1}, the smallest ACC and CON values were obtained, below 50%. For levels equal or above 100 mg L^{-1}, no classification errors were observed and both precision FOM were estimated as 100%.

The results described above indicate the lack of precision of the method at DEG concentration below 100 mg L^{-1}. This reinforces the need to establish quantitative limits for this qualitative method through the estimate of proper FOM, namely detection capability (CCβ) and decision limit (CCα). These two FOM were calculated by a POD curve estimated by a logistic fit with the outputs of the PLS-DA model for DEG contaminated class (Isabel López et al., 2015; Gondim et al., 2017). This POD curve is shown in Fig. 4, and CCα and CCβ were estimated as 52 mg

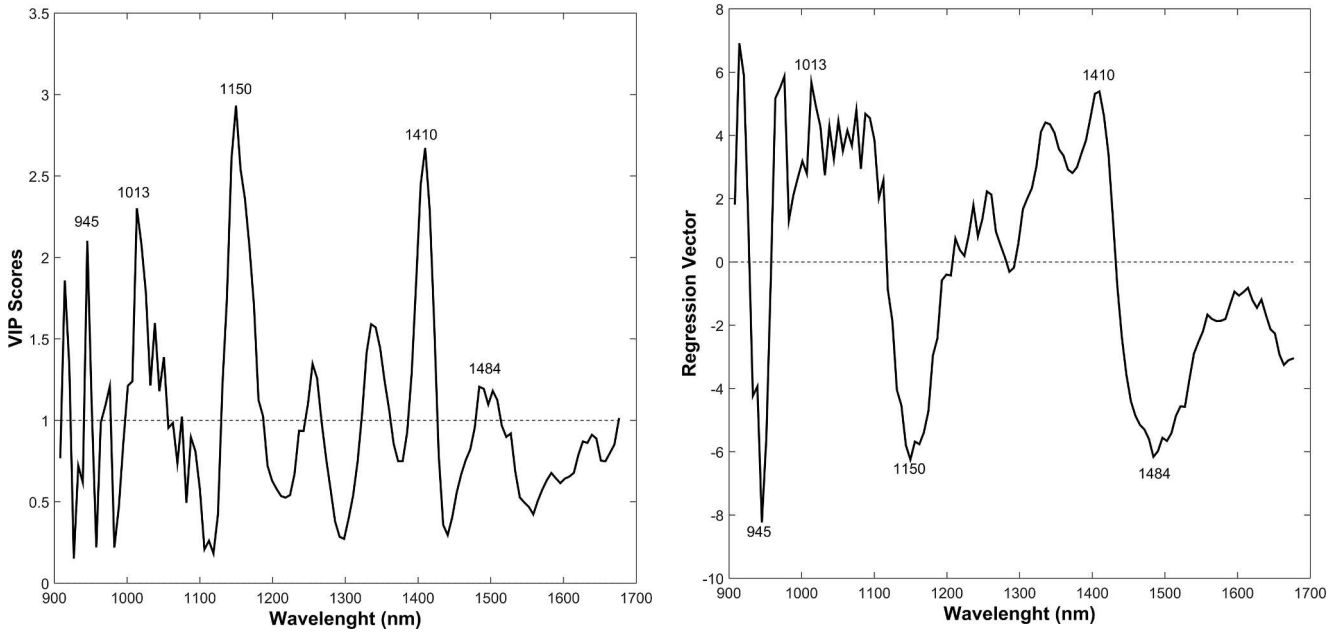

Fig. 3. Informative vectors for the PLS-DA model. (a) Variable importance in projection (VIP) scores. Black horizontal dotted line indicates the threshold of 1.0, above which variables are considered to significantly contribute to the model. (b) Regression vector.

Table 2
Estimated figures of merit for the qualitative analytical validation of the PLS-DA model. Units in %.

	FPR[a]	FNR[b]	STR[c]	SPR[d]	Accuracy	ACC[e]					CON[f]				
						50	80	100	400	1000	50	80	100	400	1000
Training	4.5	0	100	95.5	95.5	–	–	–	–	–	–	–	–	–	–
Test	6.1	0	100	93.9	93.9	44	80	100	100	100	40	81	100	100	100

[a] False positive rate.
[b] False negative rate.
[c] Sensitivity rate.
[d] Specificity rate.
[e] Accordance for concentration levels in mg L^{-1}.
[f] Concordance for concentration levels in mg L^{-1}.

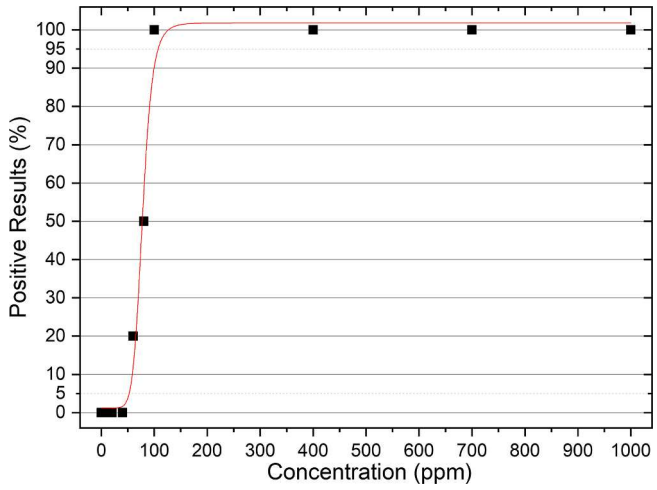

Fig. 4. Probability of detection (POD) curve obtained by logistic regression, with positive results in percentage and concentration in mg L^{-1}.

L^{-1} and 106 mg L^{-1}, respectively. CCα particularly indicates the concentration limit at which the developed method detects the contaminant with a 5% of error chance of stating that the contaminant is present when in fact it is not. Therefore, the developed method cannot be considered reliable for detecting DEG contamination in beer below 52 mg L^{-1} and its analytical range should be restricted. Naturally, the present methodology based on NIR spectroscopy and chemometrics has lower sensitivity than chromatographic methods. For example, a recently published article developed a GC–MS method for quantifying DEG in beer, which presented a limit of detection of 5.0 mg L^{-1} and a limit of quantification of 10.0 mg L^{-1} (Caldeira et al., 2021). However, our proposed alternative has several advantages and a very good cost-benefit ratio as a screening method, since it can be implemented in a portable analytical platform for on-site analysis with very reduced costs.

4. Conclusions

A simple, rapid and direct screening method for detecting the toxic diethylene glycol in beer based on a portable near-infrared spectrometer was developed. This is of special interest considering the recent occurrence in Brazil of diethylene glycol poisoning caused by beer contamination, which resulted in some deaths and dozens of intoxicated people. For the developed PLS-DA model, the decision limit (CCα) was estimated as 52 mg L^{-1}. The method was validated according to Brazilian and international guidelines (MAPA, 2014; The Commission of the European Communities, 2012), being considered accurate and precise. It was also considered robust, incorporating the variance from beers of different styles and origins that were used to build the model. The proposed

method required a small amount of sample (2 mL), spent only 30 s per spectral acquisition, did not use reagents or solvents nor generate chemical waste, and did not demand any sample preparation in addition to degassing. Finally, this methodology can be adapted to portable analytical platforms and implemented at a low cost for the quality control in brewery industry or be used as a rapid screening option for on-site forensic analysis in cases of suspected intoxication/poisoning caused by beer.

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CRediT authorship contribution statement

A.C.C. Fulgêncio: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft. **Glaucimar Alex Passos Resende:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation. **Marden Claret Fontoura Teixeira:** Data curation. **Bruno Gonçalves Botelho:** Conceptualization, Data curation, Investigation, Methodology, Resources, Supervision, Project administration, Funding acquisition. **Marcelo Martins Sena:** Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2022.133258.

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