HUNGARIAN UNIVERSITY OF
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Institute of Food Science and Technology

TAPPING INTO THE POTENTIAL OF NEAR-INFRARED SPECTROSCOPY AND AQUAPHOTOMICS FOR THE QUALITATIVE ASSESSMENT OF POWDERED FOOD MATRICES

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1. INTRODUCTION

Food powdering, a process that consists of converting food items into their powdered form, has been performed throughout history, spanning various cultures and corners of the globe. Delving into the historical contexts behind this now-considered essential unit operation entails an understanding of the drivers of powdering.

From ancient civilizations using mortar and pestle to grind spices for preservation and flavor enhancement, to the Industrial Revolution's mass production of powdered milk for convenience, the evolution of food powdering mirrors the development of human culinary ingenuity. Not only did the practical needs drive the adoption of powdered foods but the cultural and economic forces have also shaped their prominence in global cuisines. The modern era of food digitization is no different, with preferences like pivoting towards convenient over traditional home-based food and the surge of ready to eat (RTE) meals and food preparations, suited for consumers’ hectic schedules, show no sign of diminishing. However, this current of culinary trends raises a set of challenges pertaining to the safety and quality of marketed products.

More than ever, consumers now have access to a wide array of versatile powdered food items, including but not restricted to dried herbs for seasoning, finely ground coffee for instant brewing, and tomato powders for flavoring. Warranting the integrity of these marketed goods constitutes a shared challenge for all stakeholders within the food chain.

Besides qualitative degradation of powdered products, proven to dictate their marketability, adulteration is another matter of concern that poses undue risks to consumers and undermines the economy. While susceptibility to fraud is reliant upon factors such as the stringency of safety regulations, the effectiveness of traceability systems, and the vulnerability of food chains, the likelihood of being targeted is, on the other hand, inextricably linked to the economic value of the product, with a prime focus on the price-defining attributes of food matrices. Regrettably, in the attempts to uphold the desired quality, more refined adulteration methods are emerging, rendering the prevention of these malignant acts a not so trivial task. Today, perpetual fraud outbreaks and constant recalls of non-genuine products not only keep reviving authenticity concerns but also prompt the question on whether another fraud scandal would occur, and when?

To keep pace with those perpetrating food while guaranteeing prime quality, the deployment of high throughput instruments and the constant betterment of existing control systems are prerequisites. Presently used techniques encompass among many others, spectroscopic, chromatographic, DNA-based methods, etc. The rather complex nature of these conventional methods impedes their adoption as real time detection tools. Near-Infrared Spectroscopy (NIRS) has been at the forefront of adopted tools, namely since it can be less energy-intensive while still offering a greater separation of the overtone and combination bands compared with the fundamental bands. Its potential has been ascertained through applications across the food chain. Over the past years, both instrumental and software advancements facilitated the pursuit of novel research endeavors. While miniaturization led to the incorporation of near infrared spectrometers for in-situ analysis, the shift in perception with regards to relevant spectral regions as well as the mastering of multivariate data analysis tools allowed for the analysis of new matrices for uncharted applications. In essence, such upgrades granted the opportunities to analyze highly moist and liquid agrifood products, often avoided when using conventional NIRS analysis. In this regard, aquaphotomics, a novel NIRS based technique, offers a wealth of new possibilities. By taking a holistic approach to the analysis of water rich matrices and by harnessing water's feature of mirroring the state of the studied system through induced perturbations, the method has been adopted for a panoply of applications in the food industry (Atanassova et al., 2016; Gowen et al., 2012; Muncan et al., 2021; Vanoli et al., 2018; Yang et al., 2020).

Our proposed approach is a simple screening alternative requiring neither complex extraction procedures nor the use of chemicals (organic solvents), which offers reduced sampling errors compared to conventional methods. It entails shifting from the arduous determination of multiple properties to low-cost, short-time analysis where the spectral fingerprint can suffice for the characterization of samples and can be a descriptor of both the authenticity and qualitative status of the studied matrix.

2. OBJECTIVES

Seeking to extend the scope of application of aquaphotomics in food safety and quality assessment, our primary goals consisted of:

- Determining the feasibility of applying NIRS and aquaphotomics to pinpoint the admixing of Arabica coffee with inferior quality grade Robusta, both in powder and liquid form by:

1. Building qualitative models for the varietal and geographical discrimination of the Arabica and Robusta coffee types and the classification of their blends by their level of adulteration.
2. Building quantitative models for the estimation of the mixing ratio of the Arabica and Robusta coffee types.
3. Determining the water spectral pattern of the tested Arabica and Robusta coffee types and their respective aqueous blends.

- Assessing the applicability of NIRS and aquaphotomics for the detection of tomato powders adulteration by coloring and bulking agents, when presented in liquid form by:

1. Building qualitative models for the discrimination of single adulterated (coloring agent or bulking agent) and dual adulterated (coloring agent and bulking agent) mixtures by levels of adulteration.
2. Building quantitative models for the prediction of added adulterant(s)
3. Deciphering any subtle changes caused by the mere addition of the adulterants on the water spectral pattern of tomato extracts.

- Investigating the adequacy of NIRS and aquaphotomics in terms of revealing the effect of preservation processes on the aqueous extracts of selected herbs by:

1. Building qualitative models for the discrimination of the herbs according to the preservation method.
2. Building quantitative models for the estimation of the total polyphenols content (TPC) and total antioxidant capacity (TAC) in the herbal extracts

2. MATERIALS AND METHODS

2.1. Aquaphotomics for detecting adulteration in tomato extracts

For the purpose of this study, three tomato varieties (Navrongo (NA), Tytanium (TY), and Tuobodom (TU)) and three adulterants (food colorant (FC), annatto powder (AP) and corn flour (CF)), all procured from Ghanaian markets, were considered. Both single and dual adulteration mixtures were prepared. For dual adulteration, either one of the bulking agents (AP or CF) was paired with the food colorant (FC), to account for any color degradation. The adulterants were added in concentrations of 0,5%, 1%, 2%, 5%, 10%, 15% up to 20% w/w. A 10 times dilution with MilliQ water was performed and followed with a syringe-filtration through a 125 mm diameter filter paper. The obtained extracts were then subjected to spectral acquisition using the XDS Rapid Liquid Analyzer operating in transmission mode and covering the range 400-2500 nm (Figure 1).

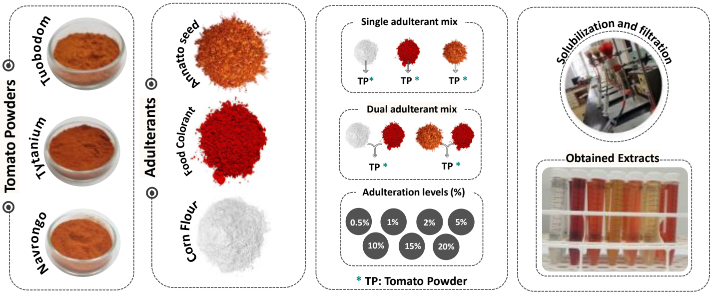

Figure 1: Authentic tomato powders, adulterants, their combinations, and aqueous extracts

The obtained spectra were then processed using R software v 4.0.2, namely aquap2 package.

To minimize spectral variability and extract any relevant patterns, a set of preprocessing tools were adopted. These consisted of smoothing (Savitzky Golay filter (2nd order polynomial) with adjustable window width (17, 19, 21, 23, 27, 29 points)), which was performed singularly or jointly with multiplicative scatter correction (MSC), detrend (DeTr), standard normal variate (SNV), 1st or 2nd derivative (Barnes et al., 1989; Savitzky & Golay, 1964).

Chemometric tools were then used to derive quantitative and qualitative models. Exploratory Principal Component Analysis (PCA) served for pattern recoginition according to the adulteration type (single or dual) and the level of adulteration. PCA based linear discriminant analyis (PCA-LDA) was then used to build classification models according to both the mixture type and extent of adulteration. Partial least squares regression (PLSR) was performed to estimate the concentration of added adulterants.

PCA-LDA and PLSR models were respectively validated by three-fold and leaving one group out cross-validations. The samples left out during PLS modelling spanned various groupings, considering criteria such as replicates, the sample types, the adulteration levels as well as the type of adulterants. Metrics such as coefficients of determination (R^2) and root mean squared errors (RMSE) were assessed, both during the calibration and cross-validation stages, for the determination of the efficiency of the models. Aquagrams were equally evaluated for any peculiar water spectral pattern of the different samples.

2.2 Aquaphotomics for coffee quality grade determination

For the purpose of this study, Arabica beans originating from Brazil (ARA1), Columbia (ARA2), and Ethiopia (ARA3) and Robusta beans sourced from Vietnam (ROB1), Uganda (ROB2), and India (ROB3) were French-roasted, ground, and procured by Bourbon café (Tahitótfalu, Hungary). Mixtures comprising 0.5, 1, 2, 3, 5, 10, 20, 35% w/w Robusta were prepared by pairing ARA3 and ROB3. Alongside these mixtures, marketed blends of different provenance and serving as test samples were considered. The 1st blend B10% consisted of 90% Arabica (South American) and 10% Robusta (South-east Asian), whereas the 2nd blend B30% comprised 70% Arabica (Central and South American) and 30% Robusta (Southeast Asian). Triplicate samples were prepared for each of the mixture concentration levels.

Water extracts of the pure coffee varieties, the mixtures, as well as the marketed blends were prepared by pouring 100 mL Milli-Q water, heated at boiling point, onto 8 g of coffee. After five minutes, the samples were filtered using a 25 µm pore-sized quantitative filter Whatman paper. The obtained extracts were cooled to room temperature (25 °C) prior to analysis (Figure 2).

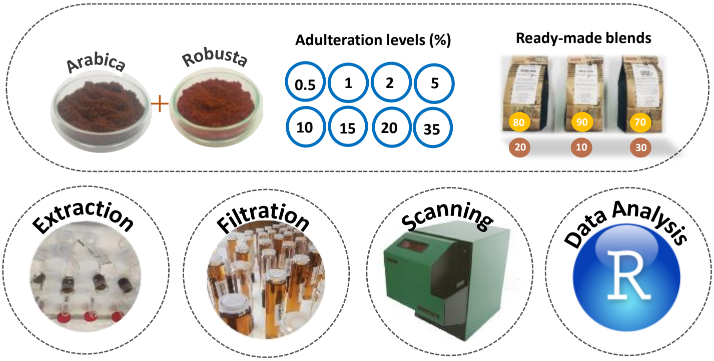

Figure 2: Schematic representation of samples preparation and analysis

A benchtop MetriNIR Spectrometer (MetriNIR Research, Development and Service Co., Budapest, Hungary) was used to collect the spectral data in the wavelength range of 740–1700 nm. Two modes of spectra acquisition were adopted, diffuse reflectance mode in the case of powders and transflectance mode for liquids.

An initial step consisted in smoothing the spectra through fitting the points into a 2nd polynomial, using Savitzky Golay filter, while also testing several window widths (11, 17, 19points). Smoothing was equally combined with other preprocessing tools such as detrend (DeTr), standard normal variate (SNV), multiplicative scatter correction (MSC) and derivatives (1st or 2nd).

Firstly, to have an overview of systematic trends, data compression was ensured by applying principal component analysis (PCA),based on the coffee variety (in case of pure varieties) and on the level of adulteration (in case of blends). These same class variables were adopted for the determination of the class memberships of the samples by PCA-LDA. These classification models were 3-fold cross-validated. Subsequently, to relate the NIR spectrum to the added Robusta, partial least squares regression modelling (PLSR) was performed. Testing the accuracy of the predictive models was ensured using the two commercialized blends B10% and B30%.

To display the water spectral pattern of the aqueous pure coffees and their corresponding blends, aquagrams were examined at both the 1st and 2nd overtone ranges, more specifically at the selected water matrix coordinates (WAMACs) of 1342, 1364, 1374, 1384, 1412, 1426, 1440, 1452, 1462, 1476, 1488, and 1512 nm (1st overtone) and at 890, 908, 924, 946, 954, 975, 1001, 1019, 1036, 1044, and 1060 nm (2nd overtone).

2.3. Aquaphotomics for tracking herbs preservation processes

For the purpose of this study, two sets of herbs, subjected to different preservation methods were tested. The first set, consisting of rosemary, marjoram and thyme, was preserved by sun-drying, shade-drying, oven-drying (40°C and 60°C), lyophilization, microwave drying (250 W and 700 W), slow and fast freezing.

The second set of herbs comprised peppermint, lovage, lemon balm, Greek oregano, tarragon, garden sage, summer savory and sweet basil. This selection was preserved by lyophilization, oven-drying (40°C and 60°C), microwave drying (250 W and 700 W), and slow freezing.

All plant stands were established in Soroksar (Pest County, Hungary) in the research field belonging to the medicinal and aromatic plants department.

After preservation, all herbs were pulverized and sieved for the obtention of powders with similar particle size. To obtain the aqueous herbal extracts, 100ml distilled and boiling water was poured over 1g of the previously ground plant material. After being soaked for 24 hours, the extracts were filtered through a 125 mm diameter filter paper (Figure 3).

For each sample type, three replicates were prepared resulting in a total of 81 samples (3 herbs × 9 preservation methods × 3 repeats) for the first set of herbs and 168 samples (8 herbs × 7 preservation methods × 3 repeats) for the second.

Near infrared and aquaphoromics based analysis was conducted at the department of food measurements and process control whereas reference measurements were conducted at the department of medicinal and aromatic plants. The latter consisted in the determination of total polyphenols content (TPC), total antioxidants capacity (TAC) and the dry matter content.

The ground herbs were scanned in reflectance mode using the benchtop XDS Rapid Content Analyzer (XDS-RCA, Metrohm, Herisau, Switzerland) while the transmission-based module of the XDS NIR spectrometer was used in case of aqueous herbal extracts.

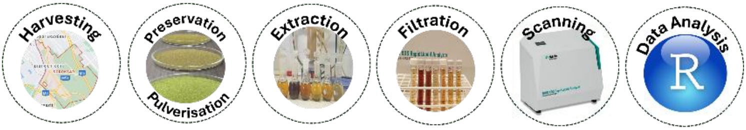

Figure 3: Schematic representation of the herbs samples preparation and analysis

Initially, spectral inspection was conducted at ranges of interest from an aquaphotomics point of view, particularly at 800-1100 nm and 1300-1600nm. A selection of spectral pretreatments was applied and their effect on the generated results was investigated. Savitzky-Golay smoothing ensured the fitting of the spectral points into a 2nd polynomial within varied window widths (21, 23, 27, 29, 31, 33). Smoothing was conducted both individually and in combination with other preprocessing tools such as 1st derivative, to resolve any overlapping peaks. Next, principal component analysis (PCA) was performed for visualization of any pattern depending on the preservation method and was followed by PCA-LDA for the classification of the ground and aqueous extracts according to the applied preservation.

Eventually, PLSR models were built to estimate the content of total polyphenols (TPC) and total antioxidant capacity (TAC). Leave one group out cross-validation based on the grouping defined by specific class variables (repeats, consecutive scans) were considered before deciding upon the model that ensures the most accurate prediction. The contributing wavelengths were inspected and their assignments were determined.

3. RESULTS

3.1. Aquaphotomics for coffee quality grade determination

An initial assessment of the ground Arabica (ARA1(Brazil), ARA2(Columbia) and ARA3(Ethiopia)) and Robusta (ROB1(Vietnam), ROB2(Uganda), and ROB3 (India)) by means of PCA-LDA modelling validated the efficiency of conventional NIR spectroscopy for the varietal and geographical discrimination of coffee with 100% correct accuracy. Using PLSR at the first overtone range (1300-1600nm), the Robusta-to-Arabica ratio in ground mixtures could be predicted with R2CV of 0.99 and an RMSECV of 2.4%. The efficacy of the implemented approach was further corroborated when marketed blends were examined, correctly discriminated, and their Robusta contents accurately estimated, the corresponding R2CV and RMSECV amounted to 0.97 and 3.93%.

The aquaphotomics based analysis of the aqueous Arabica and Robusta coffees proved that 100% authentic Robusta coffee extracts were majorly characterized by water molecules that are structured into water shells (908 nm), V1 and V2 bonded water while authentic Arabica has high hydrogen bonded water structures (1060 nm) and is rich in water clusters with two, three, and four hydrogen bonds (1018, 1036, and 1044 nm). Such results prove that a peculiar water spectral pattern (WASP) can be assigned to the pure aqueous coffee samples depending on the variety (Arabica or Robusta), with one having less hydrogen bonded water than the other. The aquagrams of the aqueous blends of ARA3 and ROB3, obtained by adding ROB3 to ARA3 in concentrations of 0.5, 1, 2, 3, 5, 10, 20, 35% w/w demonstrated that the mixing ratio can be translated in the gradual increase of certain water conformations and the decrease of other water structures. These spectral features consisted of an abundance of free water structures in Robusta-rich coffee whereas blends containing higher arabica content were characterized by high hydrogen bonded water.

As per the quantitative prediction of the added robusta in the aqueous blends of ARA3 and ROB3 by Partial least squares regression (PLSR), we proved that the NIR-based spectral analysis of these two different quality grades of coffee can accurately predict the added robusta ratio with R2 equal to 0.95 and RMSECV of 6.35% w/w. Once again, including the marketed blends resulted in a decrease in the accuracy of prediction to reach an R2CV of 0.90 and an RMSECV of 3.31%. Bands corresponding to water molecules with 2 hydrogen bonds (S2), O-H stretch second overtone of sugars and oxalic acid were the ones contributing to the obtained predictive model.

3.2. Aquaphotomics for detecting adulteration in tomato extracts

By subjecting aqueous extracts of three different tomato powder varieties (Tytanium, Tuobodom and Navrongo) comprising 0.5%, 1%, 2%, 5%, 10%, 15% up to 20% w/w of single adulterants [bulking agent (corn flour CF) and coloring agents (food colorant FC and annatto powder AP)] or their combination [(CF+FC) or (AP+FC)] to spectral screening using the benchtop XDS rapid liquid analyzer we proved that PCA-LDA classification models can accurately distinguish the samples according to their adulteration level with average recognition and prediction accuracies ranging from [97.76%-100%] and [88.59%-100%], depending on the tomato variety and the studied adulterant. As per the PLSR predictive models, they gave good estimates of the extent of adulteration, with respective R2 and RMSECV values in the ranges [0.85-0.98] and [0.55 - 1.59%w/w], depending on the adulterant (CF, FC, or AP) and adulteration type (single or dual). From aquaphotomics perspective, the results have shown that, even when average performances were achieved in PLSR, e.g., when attempting to predict the added corn, the water spectral pattern as illustrated by the aquagrams highlighted the extent of adulteration, hence confirming the feasibility of relying on aquaphotomics as a complementary technique to conventional Near infrared spectroscopy (NIRS).

3.3. Aquaphotomics for tracking herbs preservation processes

In our assessment of different herb preservation techniques (Sun and shade drying, slow and fast freezing, microwave drying at 250W and 700W and oven drying at 40°C and 60°C), we could prove that each of the applied methods had a peculiar effect on Thyme, Marjoram and Rosemary that could be traced back to their spectral features. This translated into correct assignment of the aqueous extracts of Thyme, Marjoram and Rosemary to their corresponding groups according to the varying tested preservation methods. The PCA-LDA classification models yielded good recognition and prediction accuracies of [100%-98.77%], [100%-100%], and [100%-98.77%] respectively. The method was shown to be equally effective when testing a second set of herbs, consisting of Peppermint, Lovage, Lemon Balm, Greek Oregano, Tarragon, Garden Sage, Summer Savory and Sweet Basil and preserved by lyophilization, oven-drying (40°C and 60°C), microwave drying (250 W and 700 W), and slow freezing. The NIR based classification of the aqueous extracts of these herbs resulted in good recognition accuracies ranging from [80.16% -100%] while the prediction accuracies fluctuated between 74.62% and 100%, depending on the herb.

Using XDS rapid liquid analyzer in water’s 2nd overtone range, the correlation between NIR predicted total polyphenols content (TPC) and total antioxidant capacity (TAC) values and those measured using reference methods could be established based on the spectral fingerprints of aqueous Rosemary, Thyme, and Marjoram extracts. The PLSR models validated using leave-one group-out cross-validation could predict TPC values with R^2CV of 0,94 and RMSECV of 23.12 mg GAE/g d.w and could predict TAC values with R^2CV of 0.85 and RMSECV of 32.63 mg AAE/g d.w. Such correlation could also be demonstrated in the case of the second set of herbs. The PLSR models could estimate TPC values with R^2CV and RMSECV in the ranges [0.93 -0.98] and [2.76 -15.98] mg GAE/g d.w. and could predict TAC values with R^2CV and RMSECV values in the ranges [0.93-0.97] and [1.69 – 24.05] mg AAE/g d.w, respectively. The inspection of the wavelengths contributing most to the accurate prediction of TAC and TPC accentuated that those were linked to different water conformation, from less hydrogen bonded water to more ice-like clusters of highly hydrogen bonded water. These findings prove that aquaphotomics based results align, to some extent, with the physicochemical results.

4. CONCLUSIONS AND SUGGESTIONS

The first study aimed at assessing the applicability of NIRS and aquaphotomics for the detection of tomato powders adulteration by coloring and bulking agents, when presented in liquid form.The results did signal a potential for the discrimination of single adulterated (coloring agent or bulking agent) and dual adulterated (coloring agent and bulking agent) mixtures by levels of adulteration. The estimation of added adulterants could also be achieved with good accuracies, that depended on the adulterant (coloring agent or bulking agent), but also on the type of mixture (single or dual adulterated). The extent of adulteration could be refelcted in the water spectral pattern of adulterated mixtures

Enhancing the reliability of the method and ensuring the potential implementation of this approach on an industrial scale requires further investigations with the inclusion of other prominent adulterants and tomato varieties as well as addressing the complexity of accounting for other ingredients in the tested food matrix.

The next study aimed at determining the feasibility of applying NIRS and aquaphotomics to pinpoint the admixing of Arabica coffee with Robusta, both in powder and liquid form. The applied methods offered new insights into the alterations caused by the incorporation of lower grade Robusta to higher grade Arabica, on a molecular level, which was highlighted in the respective water spectral patterns of the two authentic varieties and their blends. Qualitative and quantitative models of good accuracies corroborated the adecuacy of the method, which can be considered an alternative to laborious analysis, namely when access to ground coffee is limited.

Nevertheless, ensuring the practicality in food industries would require further refining of the applied method with the inclusion of other coffee mixtures prepared under different roasting and brewing processes but also testing its efficiency with handheld devices, better adapted to industrial settings.

In the third study, the adequacy of the method in terms of revealing the effect of preservation processes on the aqueous extracts of selected herbs was tested. Qualitative models could accurately discriminate the herbs and their extracts according to the preservation method. PLSR predictive models established the correlation between the total polyphenols content (TPC) and total antioxidant capacity (TAC) determined by reference methods and those predicted based on the NIR spectral fingerprints of the extracts.

The contributing wavebands showcased the role different water structures, from less hydrogen bonded forms to more hydrogen bonded ones, in estimating the observed alterations in the bioactive compounds of the herbs.

Ascertaining the reliability of the method as a viable alternative to conventional methods would however necessitate expanding the range of herbs studied to include a more diverse selection that takes into consideration different varieties and harvesting seasons but also evaluating other herb extraction methods.

Given the substantial economic impact automated quality control could have on the industry, we presume that shifting from laboratory measurements to on-site measurements, market-based ones through incorporating aquaphotomics based analysis via the integration of handheld spectrometers in industrial settings would be valuable. The practical implications of the presented studies could be particularly promising in cases where the authentication and detection of the adulteration is not possible in powder form and only the liquid format is available (E.g., vending machines, the expansion of the Ready-to-Drink (RTD).

Tailoring future studies towards examining other matrices would substantially contribute to the enrichment of this field and help addressing prominent challenges of our days that meet the urgent needs for food integrity and quality assurance.

Across the conducted experiments, the wavelengths contributing the most to the obtainment of accurate qualitative and quantitative models were proven to fall within the overtone ranges of water, hence accentuating the feasibility of relying on the water spectral pattern as a holistic biomarker of different food matrices as opposed to the overwhelming multiplicity of food safety and quality assessment markers. The method proves to be of particular interest with regards to eliminating sources of heterogenicity when analyzing powdered samples, which were conventionally proven to impact the spectral data and influence classification and prediction accuracies (Faqeerzada et al., 2020).

5. NEW SCIENTIFIC RESULTS

The new scientific findings, herein presented, take into consideration specific instrumental setups and well determined coffee, tomato, and herb varieties. Hence, the benchtop XDS Rapid liquid Analyzer refers to the XDS-RLA spectrometer (Metrohm, Herisau, Switzerland) used with a 1 mm pathlength quartz cuvette in transmission mode while the second benchtop spectrometer corresponds to MetriNIR spectrometer (MetriNIR Research, Development and Service Co., Budapest, Hungary) used with a circular cuvette of 0.4 mm layer thickness and operating in transflectance mode.

1. When subjecting aqueous Arabica coffee from Brazil, Columbia and Ethiopia and aqueous Robusta coffee from Vietnam, Uganda, and India to spectral analysis using the MetriNIR spectrometer I proved that a peculiar water spectral pattern (WASP) can be assigned to pure aqueous coffee samples depending on the variety. The Arabica coffee features highly hydrogen-bonded water structures (1060 nm) and is rich in water clusters with two, three, and four hydrogen bonds (1018, 1036, and 1044 nm), while Robusta coffee is primarily characterized by water molecules structured into water shells (908 nm) and V1 and V2 bonded water, having less hydrogen-bonded water than Arabica coffee. In case of blending the Arabica and Robusta coffee types, the mixing ratio is translated in the corresponding WASP with the abundance of free water structures in Robusta-rich coffee whereas blends containing higher arabica content were characterized by high hydrogen bonded water conformations.
2. In the aqueous coffee mixtures comprising 0.5, 1, 2, 3, 5, 10, 20, 35% w/w Robusta coffee from India mixed with Arabica coffee from

Ethiopia, and subjected to spectral analysis using the MetriNIR spectrometer in the 1300-1600nm range, the Robusta-to-Arabica ratio could be predicted via PLSR modelling with an R^2CV of 0.95 and an RMSECV of 6.35% w/w.

3. Aqueous thyme (*Thymus vulgaris*), marjoram (*Origanum majorana*) and rosemary (*Rosmarinus officinalis*) extracts obtained by hot water extraction following preservation by sun drying, shade drying, slow freezing, fast freezing, microwave drying (250W or 700W) or oven drying (40°C or 60°C) could be classified using PCA-LDA method based on the NIR spectra acquired in the 1300-1600nm range using the XDS Rapid liquid Analyzer. Their respective recognition and prediction accuracies consisted of [100%-98.77%], [100%-100%] and [100%-98.77%], respectively.
4. Aqueous peppermint (*Mentha × piperita*), lovage (*Levisticum officinale*), lemon balm (*Melissa officinalis*), Greek oregano (*Origanum Vulgare*), tarragon (*Artemisia Dracunculus*), garden sage (*Salvia Officinalis*), summer savory (*Satureja hortensis*) and sweet basil (*Ocimum Basilicum*) extracts obtained by hot water extraction following their preservation by lyophilization, oven-drying (40°C and 60°C), microwave drying (250 W and 700 W) or slow freezing could be classified using PCA-LDA modelling of their NIR spectra acquired in the 1300-1600nm range using the XDS Rapid liquid Analyzer with average recognition and prediction rates in the ranges [80.16% -100%] and [74.62% -100%].
5. PLSR models built on the NIR spectra of aqueous extracts of peppermint (*Mentha × piperita*), lovage (*Levisticum officinale*), lemon balm (*Melissa officinalis*), Greek oregano (*Origanum Vulgare*), tarragon (*Artemisia Dracunculus*), garden sage (*Salvia

Officinalis), summer savory (*Satureja hortensis*) and sweet basil (*Ocimum Basilicum*) obtained by different preservation methods and collected by the XDS Rapid liquid Analyzer in the 1300-1600nm range could predict their total polyphenols content (TPC) with R^2 CV and RMSECV in the ranges [0.93-0.98] and [1.69-24.15] mg GAE/g d.w and could predict total antioxidant capacity (TAC) with R^2 CV and RMSECV values in the ranges [0.94-0.98] and [0.87-15.98] mg AAE/g d.w.

6. Subjecting aqueous extracts of three different tomato powder varieties (Tytanium, Tuobodom and Navrongo) comprising of 0.5%, 1%, 2%, 5%, 10%, 15% or 20% w/w of single adulterants [bulking agent (corn flour CF), coloring agents (food colorant FC or annatto powder AP)] or their combination [(CF+FC) or (AP+FC)] to spectral screening using the XDS rapid liquid analyzer in the 1300-1600nm range we proved that PCA-LDA models can accurately classify the samples according to their adulteration level with average recognition and prediction accuracies ranging from [97.76%-100%] and [88.59%-100%], depending on the tomato variety and the studied adulterant.
7. PLSR models built on the NIR spectra of aqueous extracts of three different tomato powder varieties (Tytanium, Tuobodom and Navrongo) comprising of 0.5%, 1%, 2%, 5%, 10%, 15% or 20% w/w of single or dual adulterants acquired by the XDS rapid liquid analyzer in the 1300-1600nm range could predict the added adulterant, with respective R^2 and RMSECV values in the ranges [0.85-0.98] and [0.55-1.59%w/w], depending on the adulterant (Corn flour, Food colorant, or Annatto powder) and adulteration type (single or dual).

LIST OF PUBLICATIONS IN THE FIELD OF STUDIES

Journal articles

Bodor, Z., Benedek, C., **Aouadi, B.**, Zsom-Muha, V., & Kovacs, Z. (2022). Revealing the Effect of Heat Treatment on the Spectral Pattern of Unifloral Honeys Using Aquaphotomics. Molecules, https://doi.org/10.3390/molecules27030780. Q1 - IF 4.927

Aouadi, B.; Zaukuu, J.-L.Z.; Vitális, F.; Bodor, Z.; Fehér, O.; Gillay, Z.; Bazar, G.; Kovacs, Z. (2020). Historical Evolution and Food Control Achievements of Near Infrared Spectroscopy, Electronic Nose, and Electronic Tongue—Critical Overview. *Sensors*, https://doi.org/10.3390/s20195479. Q1 - IF 3.847

Aouadi, B.; Vitalis, F.; Bodor, Z.; Zinia Zaukuu, J.-L.; Kertesz, I.; Kovacs, Z (2022). NIRS and Aquaphotomics Trace Robusta-to-Arabica Ratio in Liquid Coffee Blends. Molecules, https://doi.org/10.3390/molecules27020388. Q1 - IF 4.927

Aouadi, B.; Laryea, D.; Aguinaga Bósquez, J.P.; Majadi, M.; Kertész, I.; Bodor, Z., Zaukuu, J.-L.Z.; Kovacs, Z.(2024). Aquaphotomics based screening of tomato powder extracts reveals susceptibility to bulking and coloring agents. Food Control, https://doi.org/10.1016/j.foodcont.2023.110163. Q1 - IF 6

Book chapters

Muncan, J; Aouadi, B; Tsenkova, R. (2024). Potential of Aquaphotomics for Determination of Quality of Natural Foods, In Megh R. Goyal, Arijit Nath, & Zoltán Kovács, (Eds.), Sustainable and Functional Foods from Plants (Chapter 12, pp.371-420). Publisher: Apple Academic Press, Inc. Co-published with CRC Press (Taylor & Francis)

Muncan, J; Aouadi, B; Vitalis, F; Kovacs, Z.; Tsenkova, R. (2020). Soil Aquaphotomics for Understanding Soil–Health Relation through Water–

Light Interaction, In: Lal, Rattan (ed.) The Soil–Human Health Nexus (Chapter 10, pp. 197-221). Publisher: CRC Press

References

Atanassova, S., Naydenova, N., Kolev, T., Iliev, T., & Mihaylova, G. (2016). Near infrared spectroscopy and aquaphotomics for monitoring changes during yellow cheese ripening. *Agric. Science Technol.*, *3*(4), 390. http://www.scopus.com/inward/record.url?eid=2-s2.0-84886778279&partnerID=tZOtx3y1

Faqeerzada, M. A., Lohumi, S., Joshi, R., & Kim, M. S. (2020). Non-Targeted Detection of Adulterants in Almond Powder Using Spectroscopic Techniques Combined with Chemometrics. *Foods*.

Gowen, A. A., Tsenkova, R., Bruen, M., & Odonnell, C. (2012). Vibrational spectroscopy for analysis of water for human use and in aquatic ecosystems. *Critical Reviews in Environmental Science and Technology*, *42*(23), 2546–2573. https://doi.org/10.1080/10643389.2011.592758

Muncan, J., Tei, K., & Tsenkova, R. (2021). Real-time monitoring of yogurt fermentation process by aquaphotomics near-infrared spectroscopy. *Sensors (Switzerland)*, *21*(1), 1–18. https://doi.org/10.3390/s21010177

Vanoli, M., Lovati, F., Grassi, M., Buccheri, M., Zanella, A., Cattaneo, T. M. P., & Rizzolo, A. (2018). Water spectral pattern as a marker for studying apple sensory texture. *Advances in Horticultural Science*, *32*(3), 343–351. https://doi.org/10.13128/ahs-22380

Yang, X., Guang, P., Xu, G., Zhu, S., Chen, Z., & Huang, F. (2020). Manuka honey adulteration detection based on near-infrared spectroscopy combined with aquaphotomics. *Lwt*, *132*(June), 109837. https://doi.org/10.1016/j.lwt.2020.109837