

Short thesis for the degree of doctor of philosophy (PhD)

**A comprehensive assessment of the environmental
indication potential of food products through
advanced analytical methods**

by **Ágota Zsófia Ragyák**

Supervisor: Dr. Edina Baranyai



UNIVERSITY OF DEBRECEN

Doctoral School of Chemistry

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I. Introduction and Objectives

The close relationship between the environment and human health has long been a central focus of scientific inquiry. However, the dynamic expansion of industrial activity, the technological transformation of agricultural production systems, and the mutually reinforcing processes of global climate change have generated new challenges over the past decades. These phenomena prompt not only localized environmental changes but also reshape ecosystem functioning on a global scale, exerting both direct and indirect effects on human nutrition and health.

A wide range of approaches is available for monitoring and assessing environmental change, among which the application of bioindicators holds particular significance. Bioindicators are organisms or groups of organisms that respond sensitively to environmental factors—such as pollution, climatic shifts, or habitat alteration—and thus provide reliable signals of ecosystem status. Traditionally, various living systems have been regarded as bioindicators; however, in recent decades, food products have also gained attention as potential indicators of environmental conditions.

Our research was grounded in the assumption that the chemical composition of certain foods may bear the imprint of environmental pressures, production processes, and geographical origin. When examined through integrated analytical and statistical methodologies, such foods may serve as indicators for tracking environmentally and medically relevant impacts. In my doctoral work, we selected three complementary model food groups for this purpose: sweeteners, honeys of different geographical origins, and Tokaj aszú wines. These products share key characteristics: they are of plant origin, shelf-stable, and possess direct or indirect links to environmental conditions. The overarching aim of the dissertation was to explore the extent to which these diverse and compositionally complex food matrices are suitable for indicating environmental influences and processing-related factors.

We set out to characterize the elemental composition of various artificial and plant-based sweeteners producer through physical or chemical transformation processes using ICP–OES measurements, to determine their

biogenic carbon content through AMS analyses, and to assess their C-stable isotope composition via IRMS, while mapping their spectral profiles using FTIR spectroscopy. Our aim was to develop a unified sample preparation protocol applicable in routine analytics for sugar alternatives of diverse composition and physical properties. Through PLS regression, we sought to construct a predictive model based on IR spectral data for estimating biocomponent content, thereby contributing to the development of cost- and time-efficient prescreening methods to support sweetener origin authentication.

Building on our research group's previous honey studies conducted in Hungary¹, we aimed to extend the work to an international level by analyzing honey collections from North America and Balkan countries. Our objective was to conduct a comprehensive chemical and isotopic characterization of the largest North American honey dataset described in the literature. Using elemental analysis (ICP–OES, ICP–MS), we planned to map the micro-, macro-, and toxic element contents of the samples, while stable isotope studies (IRMS, MC-ICP–MS) were intended to reveal information concerning geographical origin and anthropogenic influences. We also aimed to quantitatively determine the micro- and macroelement content of Serbian, Albanian, and Greek honeys (via MP-AES) and evaluate their spectral profiles using FTIR measurements. Statistical methods were employed to examine the discrimination of honeys based on botanical and geographical origin.

Drawing on our research group's previous investigations of aged honeys, we hypothesized that the complex chemical and spectral profiles of certain wines might sensitively reflect the ecological and climatic conditions of grape cultivation, thereby potentially encoding information about different vintages. To explore this, we evaluated the elemental and spectral composition of aszú samples produced in different years using ICP–OES and FTIR techniques. A central aim of my doctoral research was to uncover relationships among temporal variation, terroir-related factors, and environmental influences,

¹ Sajtos Z. et al. Rape, sunflower and forest honeys for long term environmental monitoring: Presence of indicator elements and non photosynthetic carbon in old Hungarian samples. *Sci Total Environ.* 2022;808:152044. doi:10.1016/j.scitotenv.2021.152044

as well as to evaluate the scientific robustness and interpretability of these relationships.

Our overarching hypothesis was that the shared characteristics of the examined food groups would enable us to draw generalized conclusions concerning the potential of food products to serve as indicators of environmental and human health–related factors.

II. Applied Methods

Sample collections

In assembling the sweetener collection, a primary consideration was to include natural and artificial products of various price categories and physical characteristics, reflecting consumer preferences. Priority was given to sugar alternatives that meet specific dietary requirements, thereby representing a broad spectrum of market availability. Throughout the study, we systematically sought plant-based products carrying organic certification to support subsequent comparative analyses. The resulting sample set comprises 90 sweeteners belonging to 25 different types, including both commonly used and more specialized alternatives.

Within an international collaboration, 121 honey samples from the eastern United States were obtained for analysis. Of these, 109 samples were associated with 12 states ($N \geq 3$), and 30 samples were traceable to 6 counties ($N \geq 3$). Owing to the high diversity of the honeys, only 69 samples could be unequivocally classified into one of seven botanical types ($N \geq 3$). From the Balkan region, we were able to analyze 40 Albanian, 19 Serbian, and 7 Greek honeys, each also characterized by diverse botanical origins. All honey samples were stored at room temperature in a dark, dry environment until sample preparation.

The potential of wines for environmental reconstruction was examined using 21 Tokaj aszú wines bottled annually between 1999 and 2019. This dataset represents the longest temporal sequence of aszú-type wines whose composition has been investigated to date in the scientific literature.

Sample preparation and the used analytical methods

For elemental analyses, the samples were subjected to atmospheric wet digestion using a mixture of nitric acid and hydrogen peroxide. Elemental concentrations were determined using MP-AES (Agilent Technologies 4200)

for the Balkan honey samples, ICP–OES (Agilent Technologies 5110) and ICP–MS (Agilent Technologies 8800) for the North American honeys, and ICP–OES for the sweetener and wine samples.

Through a collaboration with colleagues at the HUN-REN Institute for Nuclear Research, we conducted isotope ratio analyses on the samples. The stable carbon isotope ratios of the North American honeys were measured using IRMS (Thermo Fisher Scientific Finnigan DELTAPLUS XP). For the sweeteners, in addition to determining the $^{13}\text{C}/^{12}\text{C}$ ratio, we also quantified the radiocarbon ($^{14}\text{C}/^{12}\text{C}$) isotope ratio using AMS (MICADAS – MIni radioCARbon DAting System). For these analyses, the samples were combusted in sealed, evacuated glass tubes in the presence of MnO_2 as an oxygen source. The resulting CO_2 was purified and separated from contaminants in a vacuum line. A portion of the gas was converted to graphite for radiocarbon measurement, while the remaining fraction was used for determining the $^{13}\text{C}/^{12}\text{C}$ ratio.

Lead ($^{206}\text{Pb}/^{207}\text{Pb}$, $^{208}\text{Pb}/^{206}\text{Pb}$) and strontium ($^{87}\text{Sr}/^{86}\text{Sr}$) isotope ratios in the American honey samples were analyzed using MC-ICP–MS (Thermo Fisher Scientific Neptune Plus). During sample preparation, honey samples were digested via microwave-assisted mineralization using concentrated nitric acid. Pb and Sr were isolated from the digests using column chromatography with Pb- and Sr-specific resins.

The infrared spectra of the sweetener, Balkan honey, and aszú wine samples were recorded using an FTIR spectrometer (Agilent Technologies Cary 630) and MicroLab PC software. Following homogenization, samples were applied directly onto the ATR crystal and measured without chemical reagents. Spectra were collected in the mid-infrared region ($4000\text{--}650\text{ cm}^{-1}$) at a spectral resolution of 4 cm^{-1} .

Statistical analyses of the results were performed using The Unscrambler X, SPSS, and Past software packages.

III. New Scientific Results

1. Findings of the Sweetener Analyses

1.1 We developed a sample preparation method that can be readily integrated into routine analytics for the uniform, reagent- and cost-efficient digestion of sweeteners for elemental determinations.

Sweeteners possess complex and highly variable matrices, making their uniform, cost-effective, and time-efficient complete digestion a challenge for service laboratories handling large sample numbers. In this study, we performed microwave-assisted and atmospheric pressure digestion using various reagent ratios. Preliminary tests showed that the most effective protocol was an atmospheric pressure procedure that ensured complete oxidation of organic components with moderate reagent consumption, using a 3:1 mixture of 65% (m/m) HNO_3 and 30% (m/m) H_2O_2 . The accuracy of sample preparation and measurement was verified using an internal standard method; the recoveries of Y and Sc were within 5%, confirming the suitability of the protocol for subsequent analyses.

1.2 By examining the most comprehensive sweetener collection reported to date, we demonstrated that natural, artificial, and highly processed products can be differentiated based on their elemental composition.

Using ICP-OES, we assessed the qualitative and quantitative distribution of 20 micro- and macroelements in 90 samples. Our results showed that the 25 types of sugar alternatives differ markedly in their elemental profiles: plant-based, chemically unmodified sweeteners exhibited richer mineral content compared with artificial or highly processed products. These compositional differences were further supported by PCA, CDA, and ANOVA analyses. The primary discriminating factors were the origin of the raw materials and the degree of processing, both of which strongly influenced elemental composition.

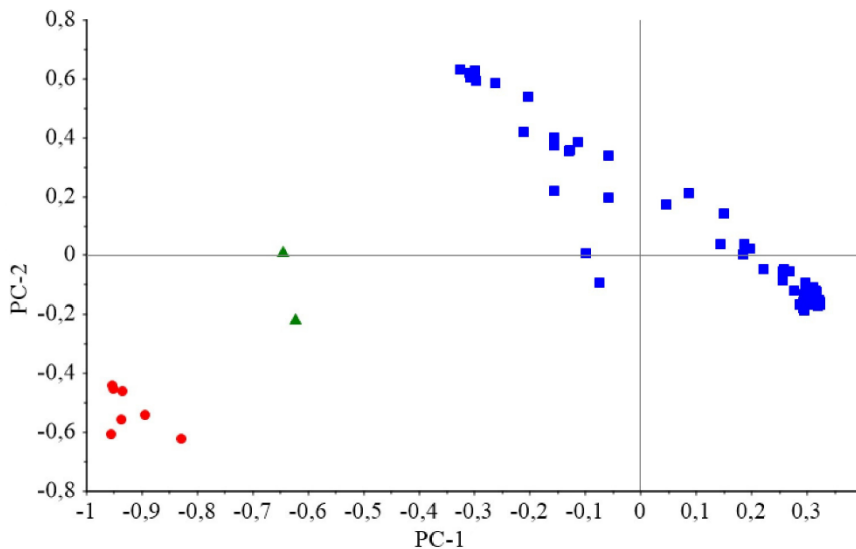
1.3 By comparing the elemental composition of organically and conventionally produced sweeteners, we showed that organic certification can influence the mineral content of plant-based sweeteners; however, these differences are not consistently observed across product types.

We analyzed organically certified and conventional variants of maple, date, agave, and rice syrups, as well as coconut blossom sugars. In date syrups, non-organic products exhibited higher K concentrations than organic ones, whereas organic coconut blossom sugars contained higher amounts of Sr, K, and Mg. For agave syrups, the non-organic sample showed elevated Al and Ba contents, while rice syrups exhibited no notable differences between the two categories. In maple syrups, ANOVA indicated significantly higher Ca concentrations ($p < 0.01$, $F = 123.94$) in organic products. Overall, we concluded that elemental differences between organic and conventional foods are product-specific and strongly dependent on cultivation conditions and processing technologies.

1.4 We demonstrated that the combined use of elemental analysis and radiocarbon measurements provides more informative characterization of sweetener samples than elemental analysis alone or elemental analysis supplemented with stable isotope ratios.

AMS measurements revealed that the lowest biobased carbon content occurred in cyclamate and saccharin mixtures, reflecting the low natural carbon content of synthetic high-intensity sweeteners. In contrast, agave, date, rice, chicory, and maple syrups, coconut blossom sugars, lucuma powder, as well as inulin and fructose samples exhibited 100% biobased carbon content, confirming their natural origin. In sweeteners composed of multiple ingredients, the cyclamate–saccharin–sucralose blend contained 27% biogenic carbon, whereas the saccharin–acesulfame-K mixture contained 55%, reflecting the proportions of the components used. When coloring the sample points in the PCA plot (based on elemental composition) according to their low (0–19%), medium (20–89%), and high (90–100%) biobased content (**Figure 1**), we

observed that biocomponent levels also reflected origin-related differences. Similarly, when the PCA was colored according to $\delta^{13}\text{C}$ values, the spatial arrangement of samples was nearly identical. However, while radiocarbon analysis provides direct quantitative information on the biobased fraction of each sample, $\delta^{13}\text{C}$ values alone do not yield explicit data regarding origin.



Biobased carbon content: ■ High ● Low ▲ Medium

Figure 1. PCA plot based on the elemental composition of the sweeteners, with samples labeled according to their biobased content. The first principal component (PC1) explains 38.9% of the total variance, while the second principal component (PC2) accounts for 63.2%.

1.5 We found that FTIR spectrum analysis can provide preliminary information on whether a sweetener's molecular pattern approaches that of a fully natural product or deviates from typical spectral profiles.

The composition of the sweeteners was analyzed using FTIR, and the resulting infrared spectra were processed using principal component analysis. Solid and liquid samples were analyzed separately, as physical state can significantly influence spectral characteristics. In both cases, synthetic sweeteners were clearly separated from natural alternatives; however, for solid sugar alternatives, the clustering was more pronounced (**Figure 2**). This phenomenon can be explained by the higher concentration of active ingredients in solid products and the relatively lower spectral contribution of excipients, which otherwise might distort the patterns. These results demonstrate that IR-based analysis can serve as a valuable, cost- and time-efficient prescreening tool for identifying spectrally distinct samples.

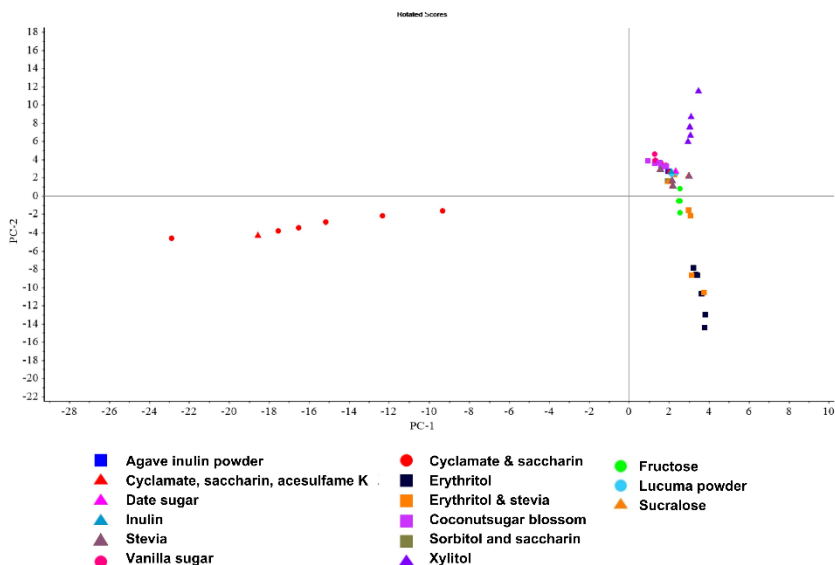


Figure 2. 2D PCA plot of IR spectra for solid sweeteners. The first principal component (PC1) explains 37.8% of the total variance, while the second principal component (PC2) accounts for 69.1%.

1.6 We developed a model to estimate the biobased carbon content of sweeteners from IR spectra using PLS regression. The resulting fit shows a strong correlation between measured and predicted values, allowing samples to be classified into low, medium, and high biobased content categories.

For the model, infrared spectral data were used as input, while biobased carbon content determined by AMS served as the reference. The full dataset was divided into a training set (one-third) and a test set (two-thirds) to validate the predictive performance. The PLS-R model (**Figure 3**) exhibited a coefficient of determination ($R^2 = 0.998$), indicating that nearly the entire variance of biobased carbon content is captured by the IR-based prediction. The regression slope (0.991) is close to the ideal value of 1, indicating minimal underestimation, while predictive accuracy remains excellent. These results show that IR spectral data can be effectively used to estimate and categorize the biobased content of sweetener samples.

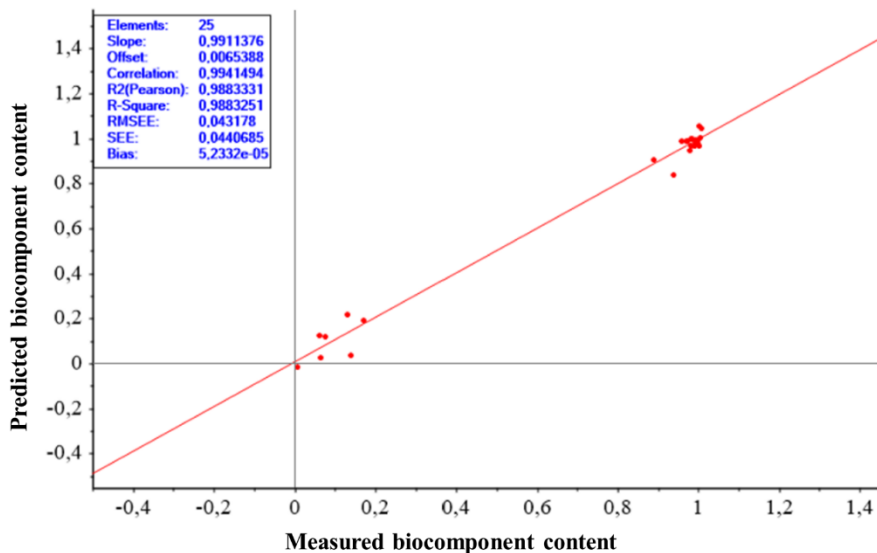


Figure 3. PLS model estimating the biobased carbon content of sweeteners based on IR spectra.

2. Findings of the Honey Analyses

2.1 We confirmed that North American honeys can be statistically distinguished based on their inorganic macro-, micro-, and toxic element content. Our results indicated that elemental profiles are more strongly influenced by botanical origin than by geographical origin.

In 121 North American honey samples, we examined the qualitative and quantitative distribution of 27 elements using ICP-MS and ICP-OES. By canonical discriminant analysis (CDA) and analysis of variance (ANOVA), we evaluated the relationship between elemental composition and botanical and geographical origin. We found that while six botanical honey types ($N \geq 3$) formed distinct clusters on the CDA biplot (**Figure 4**), the samples did not separate clearly by state of origin. When the samples were subdivided into six smaller geographic units (counties), CDA revealed clustering, suggesting that, in addition to botanical origin, the place of origin also influences composition. Overall, nectar type exerted a stronger effect on elemental content, corroborating our previous studies on Hungarian honeys.

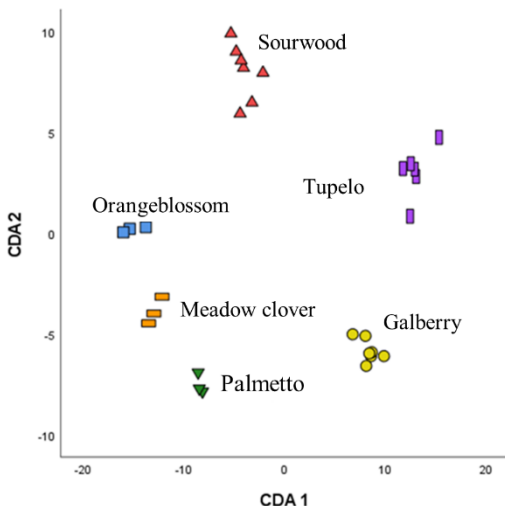


Figure 4. Classification of North American honey samples based on botanical origin.

2.2 We determined the Sr, Pb, and $\delta^{13}\text{C}$ isotope ratios of the largest number of North American honeys available in the literature. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios differed significantly between states, while $\delta^{13}\text{C}$ values indicated a region near the northern border with lower values, reflecting regional climatic effects.

MC-ICP-MS measurements revealed that Florida and the Atlantic coast exhibited lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, whereas the Appalachian region showed higher values. Statistically significant differences were observed between states, but no pronounced differences were found at the county level. This is attributed to substantial spatial variability in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios over short distances, as well as the long flight distances of bees and migratory beekeeping practices, which increase source heterogeneity. Elemental analysis detected Pb above the detection limit in four honey samples, which were further analyzed using MC-ICP-MS. The $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ ratios suggested the presence of anthropogenic Pb from fuel sources.

IRMS analyses showed that $\delta^{13}\text{C}$ values of authentic honeys ranged from -27.99‰ to -23.78‰ . Spatial distribution of $\delta^{13}\text{C}$ values did not exhibit a clear geographic pattern; however, a group of nine samples near the northern Canadian border displayed lower $\delta^{13}\text{C}$ values (below -27.05‰), likely due to higher latitude climatic conditions and canopy effects in densely forested areas, which produce ^{13}C -depleted CO_2 during plant assimilation. Overall, the $\delta^{13}\text{C}$ distribution was heterogeneous but consistent with previous international studies, confirming general uniformity in the stable isotope composition of honeys.

2.3 Using IRMS $^{13}\text{C}/^{12}\text{C}$ ratio analysis and the AOAC 978.17–1979 standard method, we identified 10 adulterated honeys among the 121 North American samples. Statistical evaluation of elemental data indicated that mineral content alone is insufficient to reliably identify adulterated products; however, it can be an effective tool when combined with other analytical methods.

The $\delta^{13}\text{C}$ values of adulterated samples exceeded -23.5% , indicating more than 13% corn-derived sugar (**Figure 5B**). This may result from supplemental corn-based feeding by beekeepers or post-processing addition of sugar syrups. Statistical analysis of elemental composition showed significant differences in Cu, Co, Cd, Rb, and Zn concentrations, but PCA did not clearly separate adulterated from authentic honeys (**Figure 5A**), suggesting that natural variability in mineral content—especially in mixed floral honeys—limits its reliability for authenticity assessment on its own.

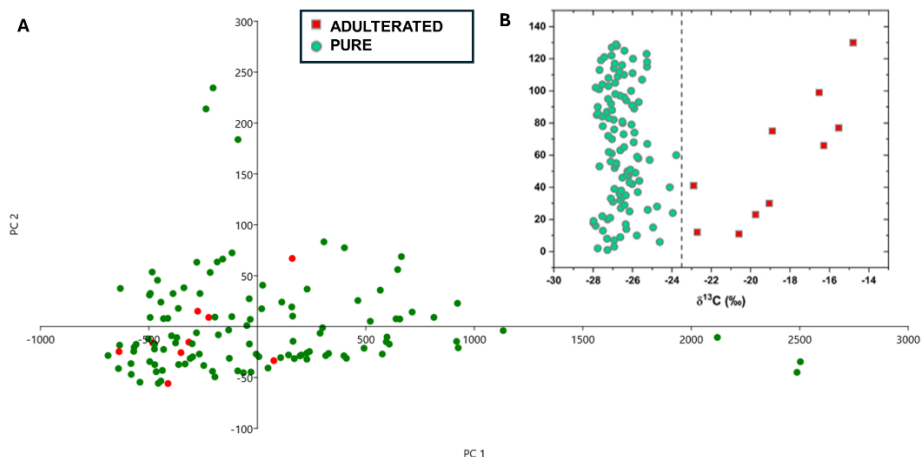


Figure 5. Analysis of authentic and adulterated honey samples. (A) PCA based on elemental composition. (B) Relative positioning based on $^{13}\text{C}/^{12}\text{C}$ ratio. Dashed line indicates the reference threshold for adulteration ($> -23.5\%$).

2.4 MP-AES analysis of Balkan honeys confirmed that Serbian, Albanian, and Greek honeys with different botanical and geographical origins can be distinguished based on elemental content. FTIR spectral analysis indicated greater fluctuations in elemental composition across geographic regions, while IR spectra within the examined range showed significant similarity.

MP-AES analysis examined 19 elements in Balkan honey samples, revealing differences in the relative proportions of micro- and macroelements among the three countries. Using CDA and ANOVA with MP-AES and FTIR data, we assessed the relationship between botanical and geographical origin and composition. CDA clearly separated Serbian and Albanian botanical honey types, while Serbian, Greek, and Albanian honeys grouped moderately but identifiable by geographical origin (**Figure 6A**). Evaluation of IR spectra in the fingerprint region ($1500\text{--}700\text{ cm}^{-1}$) showed substantial overlap among samples from different regions (**Figure 6B**).

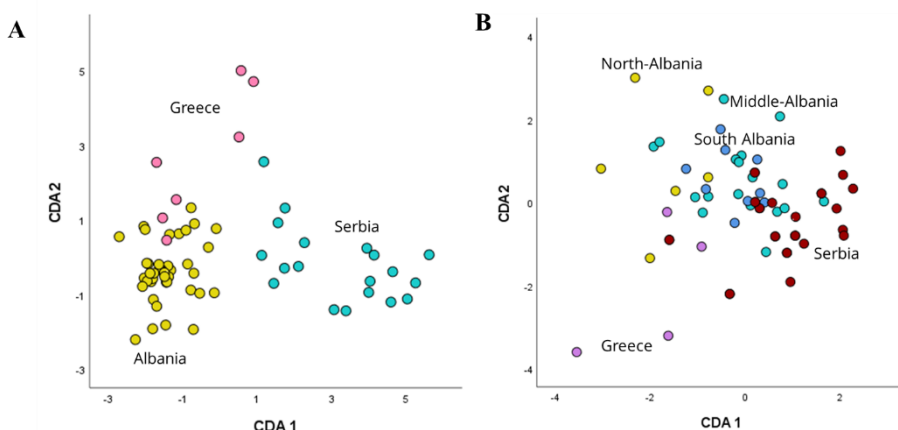


Figure 6. Classification of Serbian, Albanian, and Greek honey samples by geographical origin based on (A) elemental composition and (B) IR spectra.

3. Findings of the Aszú Wine Analyses

3.1 For the first time in the literature, we investigated the long-term potential of Tokaji aszú wines as environmental indicators. By analyzing the elemental composition of Tokaji aszú wines bottled annually between 1999 and 2019, we demonstrated that the concentrations of certain elements exhibit temporal trends. We found that temporal changes—such as variations in environmental conditions, climatic factors, or production practices—can significantly affect the metal content of the wines.

ICP-OES measurements of the aszú wines examined the qualitative and quantitative distribution of 22 elements, 17 of which were above the detection limits of the chosen analytical method. The effect of vintage on elemental composition was evaluated using multivariate statistical analysis. Samples were grouped according to vintage as follows: 1999–2005, 2006–2012, and 2013–2019 (**Figure 7A**), as well as 1999–2002, 2003–2006, 2007–2010, 2011–2014, and 2015–2019 (**Figure 7B**). CDA biplots showed clear separation between recent vintages and earlier-year groups. Evaluation of the data revealed trend-like differences in the concentrations of certain elements over time, illustrated in **Figures 7C** and **7D** for three macroelements (K, Mg, Ca) and three microelements (Sr, Ba, Cr). Potassium and chromium concentrations exhibited a strong negative correlation with vintage year (K: $p = 0.005$, $r = -0.590$; Cr: $p < 0.001$, $r = -0.707$), whereas calcium concentration showed an increasing trend over the years ($p = 0.037$, $r = 0.457$).

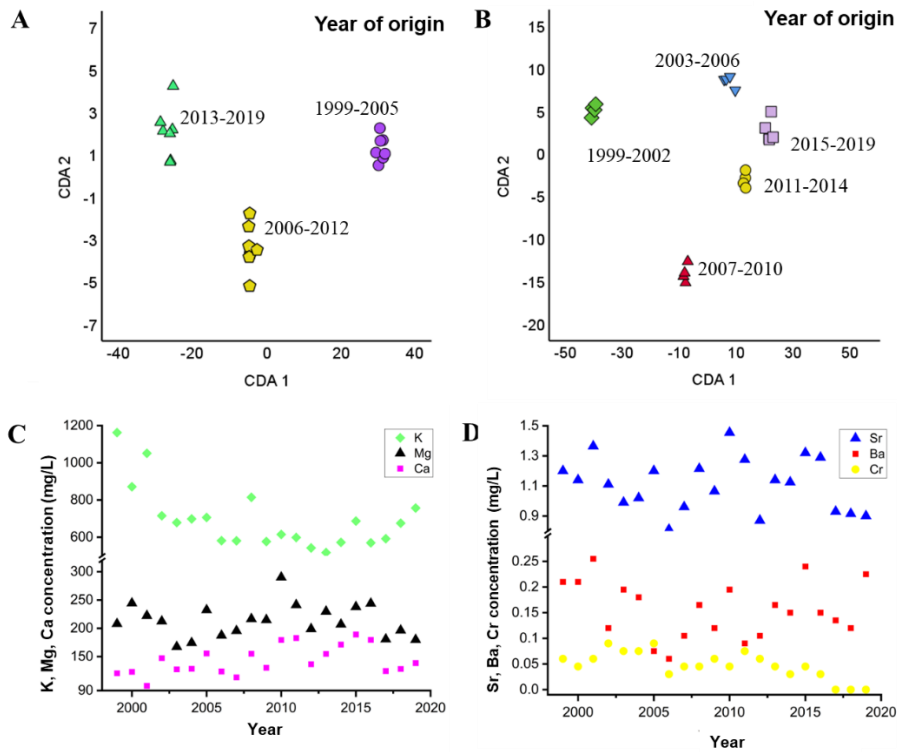


Figure 7. Investigation of the relationship between the elemental composition and vintage of Tokaji aszú wines.

3.2 We demonstrated that various meteorological factors—including annual precipitation, sunshine duration, and mean annual temperature—significantly influence the elemental content of aszú wines. Consequently, the composition of Tokaji aszú can serve as a useful proxy for environmental reconstruction studies.

3.2.1 To investigate the effect of annual precipitation on elemental composition, aszú samples were grouped according to the measured rainfall in the corresponding year: 427–456 mm, 485–551 mm, 568–640 mm, and 673–845 mm. Canonical discriminant analysis was then performed (**Figure 8A**).

Evaluation of the data revealed a positive correlation between zinc content and annual precipitation ($p = 0.036$, $r = 0.46$; **Figure 8B**). In contrast, barium content correlated negatively with the previous year's precipitation ($p = 0.020$, $r = -0.529$; **Figure 8C**), and calcium content correlated negatively with precipitation measured two years prior ($p = 0.042$, $r = -0.471$). These trends can be explained by the influence of soil management practices and rainfall characteristics on the leaching of specific elements from vineyard soils.

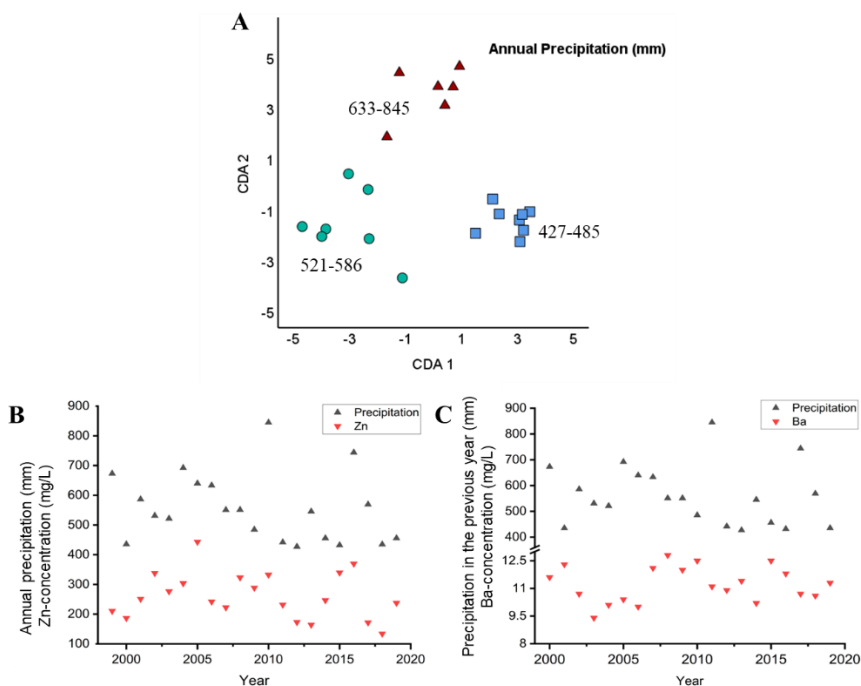


Figure 8. Investigation of the relationship between elemental composition of Tokaji aszú wines and annual precipitation.

3.2.2 We examined whether the number of annual sunshine hours affects the composition of aszú wines and, if so, to what extent. For CDA analysis, the wine samples were grouped according to the recorded sunshine hours in the given year: 1920–2089, 2121–2283, 2309–2478, and 2536–2672 hours (**Figure 9A**). The observed group separation was primarily due to changes in four elements related to sunlight exposure. Significant negative correlations were found between annual sunshine hours and the concentrations of chromium (Cr: $p = 0.001$, $r = -0.650$), strontium (Sr: $p = 0.021$, $r = -0.499$), zinc (Zn: $p = 0.026$, $r = -0.484$), and phosphorus (P: $p = 0.048$, $r = -0.437$) (**Figure 9B**). This phenomenon can be explained by plants responding to increased sunlight by enhancing photosynthetic activity and growth, which leads to redistribution of available nutrients to the plant tissues where they are most required.

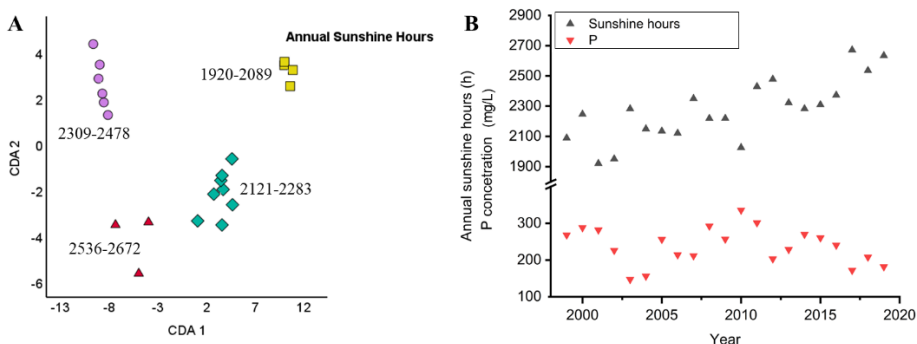


Figure 9. Investigation of the relationship between elemental composition of Tokaji aszú wines and annual sunshine hours.

3.2.3 To assess the effect of mean annual temperature on elemental content, wines were grouped according to the yearly average temperature: 9.8–10.7 °C, 11.1–11.5 °C, and 11.6–12.4 °C. The spatial separation of these groups in Figure 10A reflects a negative correlation between annual mean temperature and the Cr content of the samples ($p = 0.001$, $r = -0.650$; Figure 10B). This

trend can also be attributed to physiological changes in the vines under differing thermal conditions.

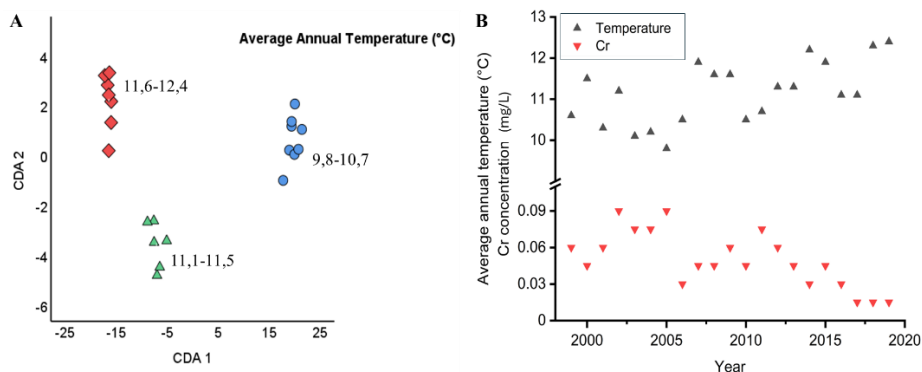


Figure 10. Investigation of the relationship between elemental composition of Tokaji aszú wines and mean annual temperature.

3.3 Our study was the first to examine the applicability of FTIR for comparative analysis of aged aszú wines. We demonstrated that FTIR can be a useful tool to explore the effects of environmental factors on wine composition and quality.

The infrared spectra of 21 wine samples were recorded using FTIR over the spectral range of $4000\text{--}650\text{ cm}^{-1}$. For statistical evaluation, the fingerprint region of $1800\text{--}600\text{ cm}^{-1}$ was used. Each wine was measured in at least three replicates, and principal component analysis was applied to visualize similarities and differences among the samples. We found that the observed visual clustering could be explained by environmental characteristics in the respective years. For example, wines bottled in 2007, 2012, 2018, and 2019 clustered closely, reflecting unusually warm summers that led to above-average berry sugar content. This is further supported by increased spectral intensity in the $1150\text{--}1000\text{ cm}^{-1}$ range, corresponding to carbohydrate-related bands.

IV. Potential Applications of the Results

During my doctoral research, the indicative potential of three different high-sugar, naturally derived food types—sweeteners, honeys, and Tokaji aszú wines—was investigated, with a particular focus on their elemental composition and isotopic ratios. Our integrated analyses aimed to contribute to the assessment of these foods as environmental indicators and to provide a scientific basis for more precise evaluation of environmental pressures and human exposure risks.

We developed a cost- and reagent-efficient atmospheric digestion protocol for elemental analysis of sweeteners, which exhibit high variability in their matrices. This method represents a practical option for service laboratories handling large sample numbers. Using this approach, we assembled the most comprehensive collection of sugar alternatives reported in the literature to date. The study presented in this dissertation—and soon to be submitted for publication—provides an extensive characterization of sweetener compositions, significantly expanding the current knowledge base. Our findings demonstrate that combining elemental and radiocarbon analyses offers an effective approach for categorizing sweeteners by origin.

Moreover, the PLS-R model we developed to estimate the biogenic carbon content of sweeteners from IR spectra is particularly useful in practice. This stable model allows routine origin verification to rely solely on IR spectral acquisition and model-based evaluation, while costly and time-consuming AMS measurements are required only for critical samples. In the long term, this approach not only enables rapid and non-destructive preliminary classification but also offers a risk-based, cost-efficient alternative for routine analysis of large sample numbers.

Honey is among the most frequently adulterated foods worldwide; thus, knowledge of the composition of honeys from different botanical and geographic origins is crucial for developing authentication and quality-control procedures. Our honey analyses contribute to expanding the international literature. Prior to our work, data on North American honey were limited; therefore, our results can serve as reference material for comparative studies of

the continent's honeys. Analysis of Serbian and Albanian honey samples provides valuable information on the region and facilitates comparative evaluation, which is particularly important for comprehensive chemical characterization and international contextualization of Balkan honeys. Studying samples from diverse geographic regions can support the creation of reference databases that improve the identification of adulteration practices and enable verification of the reliability of products in global trade.

Through analysis of wine samples spanning more than two decades, we were the first in the literature to demonstrate that Tokaji aszú wines can serve as bioindicators for long-term environmental reconstruction. As archival records, these wines reflect long-term trends in climatic conditions, soil use, and viticultural practices, while also providing insight into the factors determining the characteristics of individual vintages. In the long term, our findings offer critical guidance for establishing more informed and sustainable vineyard management strategies, as monitoring vintage-specific effects over decades allows prediction of the impact of environmental changes on grape and wine chemical composition. In response to challenges posed by climate change, this approach can contribute to identifying cultivation and processing practices best suited to local terroir, thereby supporting quality preservation and adaptive winemaking decisions. This is particularly valuable for distinguished wine regions like Tokaj, where the preservation of tradition and natural environment plays a key role in origin protection and maintaining cultural heritage.

List of Publications

International Peer-Reviewed Articles Forming the Basis of the Dissertation

1. Á. Ragyák, Z. Sajtos, E. Baranyai, E. László: Long term compositional profiling of historical Tokaji aszú wines. npj Science of Food 9, 101 (2025), DOI: 10.1038/s41538-025-00468-x. **IF: 7.8, D1 – Featured as “Publication of the Month” by the Hungarian Academy of Sciences, October 2025.**
2. T. Varga, Z. Sajtos, E. Baranyai, Z. Lisztes-Szabó, Á. Ragyák, M. Molnár, A.J. T. Jull, S. Szabó, K. Hajduné Kosdi, I. Futó, J. M. Kaste: Isotopic study of honey documents widespread plant uptake of old carbon in North America. Science of The Total Environment 947:174691 (2024), DOI: 10.1016/j.scitotenv.2024.174691. **IF: 8, D1**
3. Á. Ragyák, T. Varga, N.A.K. Ahmed, M. Braun, A. Horváth, Z. Lisztes-Szabó, S. Szabó, J. Kaste, C.P. Bataille, M. Reich, E. Baranyai, Z. Sajtos: Comprehensive analysis of the elemental composition and isotope ratios of honeys from US East Coast. Food Chemistry (2025), **IF: 9.8 (submitted)**

International Peer-Reviewed Articles Not Directly Related to the Dissertation

1. Á. Jakab, N. Balla, Á. Ragyák, F. Nagy, F. Kovács, Z. Sajtos, A. M. Borman, I. Pócsi, E. Baranyai, L. Majoros, R. Kovács: Transcriptional profiling of the Candida auris response to exogenous farnesol exposure. mSphere 6(5):e00710-21 (2021), DOI: 10.1128/mSphere.00710-21. **IF: 5.029, Q1**

2. Á. Jakab, F. Kovács, N. Balla, Z. Tóth, Á. Ragyák, Z. Sajtos, K. Csillag, C. Nagy-Köteles, D. Nemes, I. Bácskay, I. Pócsi, L. Majoros, Á. T. Kovács, R. Kovács: Physiological and transcriptional profiling of surfactin exerted antifungal effect against *Candida albicans*. *Biomedicine & Pharmacotherapy* 152(7):113220 (2022), DOI: 10.1016/j.biopha.2022.113220. **IF: 7.5, Q1**
3. V. Oláh, M. Irfan, Z. Szabó Barnáné, Z. Sajtos, Á. Ragyák, B. Dönczö, Marcel A. K. Jansen, S. Szabó, I. Mészáros: Species- and metal-specific responses of the ionome of three duckweed species under chromate and nickel treatments. *Plants* 12(1):180 (2023), DOI: 10.3390/plants12010180. **IF: 4.0, Q1**
4. N. Balla, Á. Jakab, F. Kovács, Á. Ragyák, Z. Tóth, D. Balázsi, L. Forgács, A. Bozó, F. Al Refai, AM. Borman, L. Majoros, R. Kovács: Total transcriptome analysis of *Candida auris* planktonic cells exposed to tyrosol. *AMB Express* 13(1) (2023), DOI: 10.1186/s13568-023-01586-z. **IF: 3.7, Q2**
5. Z. Sajtos, Á. Ragyák, F. Hódi, V. Szigeti, G. Bellér, E. Baranyai: 5-Hydroxymethylfurfural content of old honey samples – does the sticky treat really last forever? *LWT* 193:115781 (2024), DOI: 10.1016/j.lwt.2024.115781. **IF: 6.6, D1**
6. Á. Jakab, F. Kovács, N. Balla, C. Nagy-Köteles, Á. Ragyák, F. Nagy, A. Borman, L. Majoros, R. Kovács: Comparative transcriptional analysis of *Candida auris* biofilms following farnesol and tyrosol treatment. *Microbiology Spectrum* 12(4):e0227823 (2024), DOI: 10.1128/spectrum.02278-23. **IF: 3.8, Q1**
7. D. Tőzsér, D. O. Idehen, J. D. Osazuwa, J. E. Sule, Á. Ragyák, Z. Sajtos, T. Magura: Early-stage growth and elemental composition patterns of *Brassica napus* L. under Cd-Zn stress. *Chemosphere* 351:141235 (2024), DOI: 10.1016/j.chemosphere.2024.141235

8. D. Tőzsér, J.D. Osazuwa, J.S. Elias, D.O. Idehen, D.I. Gutiérrez Pérez, Á. Ragyák, Z. Sajtos, T. Magura: Comparative analysis of the short-term germination and metal accumulation patterns of two Sorghum hybrids. Environmental Geochemistry and Health 47, 178 (2025), DOI: 10.1007/s10653-025-02485-x. **IF: 3.8, Q1**

9. F. Kovács, Á. Jakab, N. Balla, Z. Tóth, D. Balázs, L. Forgács, A. Harmath, A. Bozó, Á. Ragyák, L. Majoros, R. Kovács: A comprehensive analysis of the effect of quorum-sensing molecule 3-oxo-C12-homoserine lactone on Candida auris and Candida albicans. Biofilm 9(1):100259 (2025), DOI: 10.1016/j.biofilm.2025.100259. **IF: 4.9, Q1**

International Conference Presentations and Posters Related to the Dissertation

1. Á. Ragyák, Z. Sajtos, E. László, E. Baranyai: *Elemental profiling of historical Tokaj wine samples – Exploring potential for environmental assessment*. XIX. Carpathian Basin Environmental Science Conference, Debrecen, Hungary, 2024.04.03-05 (Poster)

2. Á. Ragyák, T. Varga, N.A.K. Ahmed, M. Braun, A. Horváth, L. Palcsu, Zs. Lisztes-Szabó, M. Molnár, A.J. T. Jull, Sz. Szabó, K.H. Kosdi, I. Futó, J. Kaste, E. Baranyai, Z. Sajtos: *Elemental and isotopic analysis of aged honeys – A North American perspective on unique sweeteners*. 11th Nordic Conference on Plasma Spectrochemistry, Loen, Norway, 2024.06.09-13 (Poster)

3. Á. Ragyák, Z. Sajtos, E. László, E. Baranyai: *Elemental profiling of historical Tokaj wine samples – Exploring potential for environmental assessment*. European Winter Conference on Plasma Spectrochemistry (EWCPS), Berlin, Germany, 2025.03.02-07 (Oral)

4. Á. Ragyák, M. Csontos, T. Varga, Z. Sajtos, E. Baranyai: *Complex analytical investigation of natural and artificial sweeteners*. EWCPs, Berlin, Germany, 2025.03.02-07 (Poster)
5. Z. Sajtos, Á. Ragyák, T. Varga, N.A.K. Ahmed, M. Braun, A. Horváth, L. Palcsu, Z. Lisztes-Szabó, M. Molnár, A.J.T. Jull, S. Szabó, K. Hajduné Kosdi, I. Futó, J. Kaste, E. Baranyai: *Environmental reconstruction studies with elemental and isotopic analysis of honey samples – North American point of view*. EWCPs, Berlin, Germany, 2025.03.02-07 (Oral)
6. Z. Sajtos, Á. Ragyák, T. Varga, Z. Gajdos, Z. Lisztes-Szabó, A.J.T. Jull, M. Molnár: *Reconstructing environmental changes through elemental and isotopic analysis of honey*. 23rd International Symposium and Summer School on Bioanalysis, Zagreb, Croatia, 2025.07.06-12 (Oral)
7. Á. Ragyák, M. Csontos, T. Varga, Z. Sajtos, E. Baranyai: *Complex analytical investigation of natural and artificial sweeteners*. Euroanalysis, Barcelona, Spain, 2025.08.31-09.04 (Poster)
8. Z. Sajtos, Á. Ragyák, T. Varga, N.A.K. Ahmed, M. Braun, A. Horváth, L. Palcsu, Z. Lisztes-Szabó, M. Molnár, S. Szabó, K. Hajduné Kosdi, I. Futó, J. Kaste, E. Baranyai: *Elemental composition and isotope ratio analysis of honeys from US East Coast*. Euroanalysis, Barcelona, Spain, 2025.08.31-09.04 (Poster)

Hungarian Conference Presentations and Posters Related to the Dissertation

1. Á. Ragyák: Reconstruction of environmental changes through complex analytical investigation of honey and Tokaji aszú samples. Institutional ÚNKP Conference, University of Debrecen, Debrecen, Hungary, 29 Jan 2024 (Oral)
2. Á. Ragyák, Z. Sajtos, E. László, E. Baranyai: The imprint of environmental changes in a wine glass – Elemental analysis of Tokaji aszú wines. 63rd Hungarian Spectrochemistry Meeting, Balatonszárszó, Hungary, 6–8 Mar 2024 (Poster)
3. Á. Ragyák, M. Csontos, T. Varga, Z. Sajtos, E. Baranyai: Complex analytical investigation of natural and artificial sweeteners. 64th Hungarian Spectrochemistry Meeting, Balatonszárszó, Hungary, 5–7 Nov 2025 (Poster)
4. Z. Sajtos, Á. Ragyák, T. Varga, N. A. K. Ahmed, A. Horváth, M. Braun, Z. Lisztes-Szabó, S. Szabó, J. Kaste, C. P. Bataille, M. Reich, E. Baranyai: Environmental reconstruction studies through elemental and isotopic analysis of honey samples. 64th Hungarian Spectrochemistry Meeting, Balatonszárszó, Hungary, 5–7 Nov 2025 (Oral)
5. E. Baranyai, Á. Ragyák, Z. Sajtos, E. László: Long-term analytical investigation of Tokaji aszú wines. 64th Hungarian Spectrochemistry Meeting, Balatonszárszó, Hungary, 5–7 Nov 2025 (Oral)



Registry number:
Subject:

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PhD Publication List

Candidate: Ágota Zsófia Ragyák
Doctoral School: Doctoral School of Chemistry
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List of publications related to the dissertation

Foreign language scientific articles in international journals (2)

1. **Ragyák, Á.**, Sajtos, Z., Baranyai, E., László, E.: Long term compositional profiling of historical tokaji aszú wines.
npj Sci Food. 9 (1), 1-10, 2025. EISSN: 2396-8370.
DOI: <http://dx.doi.org/10.1038/s41538-025-00468-x>
IF: 7.8 (2024)
2. Varga, T., Sajtos, Z., Baranyai, E., Lisztes-Szabó, Z., **Ragyák, Á.**, Molnár, M., Jull, A. J. T., Szabó, S., Hajduné Kosdi, K., Futó, I., Kaste, J. M.: Isotopic study of honey documents widespread plant uptake of old carbon in North America.
Sci. Total Environ. 947, 1-11, 2024. ISSN: 0048-9697.
DOI: <http://dx.doi.org/10.1016/j.scitotenv.2024.174691>
IF: 8

List of other publications

Foreign language scientific articles in international journals (9)

3. Kovács, F., Jakab, Á., Balla, N., Tóth, Z., Balázs, D., Forgács, L., Harmath, A., Bozó, A., **Ragyák, Á.**, Majoros, L., Kovács, R. L.: A comprehensive analysis of the effect of quorum-sensing molecule 3-oxo-C12-homoserine lactone on *Candida auris* and *Candida albicans*.
Biofilm. 9, 1-12, 2025. ISSN: 2590-2075.
DOI: <http://dx.doi.org/10.1016/j.biofilm.2025.100259>
IF: 4.9 (2024)
4. Tőzsér, D., Osazuwa, J. D., Sule, J. E., Idehen, D. O., Gutiérrez Pérez, D. I., **Ragyák, Á.**, Sajtos, Z., Magura, T.: Comparative analysis of the short-term germination and metal accumulation patterns of two *Sorghum* hybrids.
Environ. Geochem. Health. 47 (5), 1-18, 2025. ISSN: 0269-4042.
DOI: <http://dx.doi.org/10.1007/s10653-025-02485-x>
IF: 3.8 (2024)





5. Jakab, Á., Kovács, F., Balla, N., Nagy-Köteles, C., **Ragyák, Á.**, Nagy, F., Borman, A. M., Majoros, L., Kovács, R. L.: Comparative transcriptional analysis of *Candida auris* biofilms following farnesol and tyrosol treatment.
Microbiol. Spectr. 12 (4), 1-12, 2024. ISSN: 2165-0497.
DOI: <https://doi.org/10.1128/spectrum.02278-23>
IF: 3.8
6. Tözsér, D., Idehen, D. O., Osazuwa, J. D., Sule, J. E., **Ragyák, Á.**, Sajtos, Z., Magura, T.: Early-stage growth and elemental composition patterns of *Brassica napus* L. in response to Cd-Zn contamination.
Chemosphere. 351, 1-10, 2024. ISSN: 0045-6535.
DOI: <http://dx.doi.org/10.1016/j.chemosphere.2024.141235>
7. Sajtos, Z., **Ragyák, Á.**, Hódi, F., Szigeti, V., Bellér, G., Baranyai, E.: Hydroxymethylfurfural content of old honey samples: Does the sticky treat really last forever?
LWT-Food Sci. Technol. 193, 1-8, 2024. ISSN: 0023-6438.
DOI: <http://dx.doi.org/10.1016/j.lwt.2024.115781>
IF: 6.6
8. Oláh, V., Irfan, M., Barnáné Szabó, Z., Sajtos, Z., **Ragyák, Á.**, Dönczö, B., Jansen, M. A. K., Szabó, S., Mészáros, I.: Species- and Metal-Specific Responses of the Ionome of Three Duckweed Species under Chromate and Nickel Treatments.
Plants-Basel. 12 (1), 1-16, 2023. ISSN: 2223-7747.
DOI: <http://dx.doi.org/10.3390/plants12010180>
IF: 4
9. Balla, N., Jakab, Á., Kovács, F., **Ragyák, Á.**, Tóth, Z., Balázs, D., Forgács, L., Bozó, A., Al Refai, F., Borman, A. M., Majoros, L., Kovács, R. L.: Total transcriptome analysis of *Candida auris* planktonic cells exposed to tyrosol.
AMB Express. 13 (1), 1-10, 2023. EISSN: 2191-0855.
DOI: <http://dx.doi.org/10.1186/s13568-023-01586-z>
IF: 3.5
10. Jakab, Á., Kovács, F., Balla, N., Tóth, Z., **Ragyák, Á.**, Sajtos, Z., Csillag, K., Nagy-Köteles, C., Nemes, D., Bácskay, I., Pócsi, I., Majoros, L., Kovács, Á. T., Kovács, R. L.: Physiological and transcriptional profiling of surfactin exerted antifungal effect against *Candida albicans*.
Biomed. Pharmacother. 152, 1-10, 2022. ISSN: 0753-3322.
DOI: <http://dx.doi.org/10.1016/j.biopha.2022.113220>
IF: 7.5
11. Jakab, Á., Balla, N., **Ragyák, Á.**, Nagy, F., Kovács, F., Sajtos, Z., Tóth, Z., Borman, A. M., Pócsi, I., Baranyai, E., Majoros, L., Kovács, R. L.: Transcriptional profiling of the *Candida auris* response to exogenous farnesol exposure.
mSphere. 6 (5), 1-12, 2021. EISSN: 2379-5042.
DOI: <http://dx.doi.org/10.1128/mSphere.00710-21>
IF: 5.029





Foreign language abstracts (2)

12. Kovács, R. L., Kovács, F., Balla, N., **Ragyák, Á.**, Sajtos, Z., Majoros, L., Jakab, Á.:
Transcriptional changes and related physiological effect caused by tyrosol exposure in
Candida auris.
In: ECFG16 abstractbook. Eds.: Universität Innsbruck, PCO Tyrol Congress, Innsbruck, 425-
426, 2023.
13. Kovács, R. L., Jakab, Á., **Ragyák, Á.**, Sajtos, Z., Nagy, F., Baranyai, E., Pócsi, I., Majoros, L.:
Transcriptomic approaches for the farnesol exposure of Candida auris.
Acta Microbiol. Immunol. Hung. 68, 85, 2021. ISSN: 1217-8950.

Total IF of journals (all publications): 54,929

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The Candidate's publication data submitted to the Tudóstér have been validated by DEENK on the basis of the Journal Citation Report (Impact Factor) database.

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