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Editorial

Editorial "High-Performance Green Extraction of Natural Products"

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There has been, to-date, a large number of studies pertaining to the exploitation of plant resources (herbs, botanicals, processing by-products) for the production of extracts enriched with bioactive substances. The interest has been focused on the development of efficient and cost-effective downstream processes, which aim at producing commodities on the basis of either crude or purified extracts.

Traditional extraction techniques, including percolation, decoction/infusion generation, maceration, etc., are currently being replaced by cutting-edge, sophisticated technologies with higher efficiency and selectivity, and a more eco-friendly profile. Advanced extraction methodologies based on ultrasonication, microwaves, pulsed electric fields, high voltage discharges, enzymes, pressurized liquids, supercritical fluids, deep eutectic solvents, etc., have in many instances proven to be more targeted, high-performing, straightforward, fast, sustainable, fully automated, and with relatively low capital cost.

This Special Issue addresses the concept of innovative and emerging strategies that aim at effectively implementing green technologies for the extraction of bioactive compounds from plant resources.

Loukri et al. [1] examined the extraction of caffeine and chlorogenic acids from coffee pulp, a by-product of coffee production, using aqueous solutions of β-cyclodextrin (β-CD) as a non-conventional solvent. The parameters of β-CD concentration, liquid-to-solid ratio, and temperature were evaluated based on the antiradical activity and the caffeine content, by deploying the response surface methodology. The sensory profiles of brews prepared with coffee and coffee pulp with or without cyclodextrin were studied with quantitative descriptive analysis. The brew from the by-product had fruity, botanic, sweet, and sour sensory properties, and cyclodextrin was found to be able to affect the overall taste of the brew.

Dabetić et al. [2] investigated the exploitation of deep eutectic solvents (DESs) (choline chloride: citric acid and choline chloride: glucose) as solvents for extracting valuable phenolic antioxidants from grapes. Investigation was conducted on ten grape varieties, observing seeds and skin as different matrices. Overall results support that DESs (particularly choline chloride: citric acid) were comparable to conventional solvents, and in most cases even outperformed acidified aqueous ethanol with regard to extraction efficiency and antioxidant activity. Regardless of varietal distinctions, grape seeds were found to have higher antioxidant capacity compared to grape skins, in accordance with their polyphenol concentrations.

Lakka et al. [3] developed a simple, straightforward, and green extraction methodology to effectively recover potato peel polyphenols, using hydroxypropyl β-cyclodextrin (HP-β-CD). After an initial assay to identify the optimal HP-β-CD concentration that would provide increased extraction yield, optimization based on response surface methodology enabled maximization of the extraction performance. Testing of temperatures higher than 30 °C and up to 80 °C did not favor higher yields. The extracts obtained with HP-β-CD were slightly richer in polyphenols than extracts prepared with conventional solvents, such as aqueous ethanol and methanol, displaying similar antioxidant characteristics. The major polyphenols that could be identified in the extracts were

neochlorogenic, chlorogenic, caffeic, and ferulic acids. The outcome of this study demonstrated that HP-β-CD may be used as a highly effective green means of recovering potato peel polyphenols, at near-ambient temperature.

In another study, Lakka et al. [4] established a green extraction process using a novel eco-friendly natural deep eutectic solvent, composed of glycerol and nicotinamide, to produce polyphenol-enriched extracts from *Moringa oleifera* leaves. Furthermore, sample ultrasonication prior to batch stirred-tank extraction was studied to examine its usefulness as a pretreatment step. Optimization of the extraction process through the response surface methodology showed that the maximum total polyphenol yield could be achieved after a 30 min ultrasonication pretreatment, but the difference from the yield obtained from the non-pretreated sample was statistically non-significant. Extraction kinetics revealed that the activation energy for the ultrasonication-pretreated samples was more energy-demanding, a fact attributed to phenomena pertaining to washing of the readily extracted polyphenols during pretreatment. Liquid chromatography-diode array-mass spectrometry showed that ultrasonication pretreatment may have a limited positive effect on polyphenol extractability, but the overall polyphenolic profile was identical for the ultrasonication-pretreated and non-pretreated samples.

Jin et al. [5] studied the application of deep eutectic solvents (DESs) as safe and efficient extraction media that could yield maximized skin-related bioactivities from a mixture of long-lived trees: *Ginkgo biloba* L., *Cinnamomum camphora* (L.) J. Presl., and *Cryptomeria japonica* (L.f.) D. Don, native to Asia. Various DESs were synthesized from cosmetics-compatible compounds and used to prepare leaf extracts. A DES containing glycerol and xylitol yielded the highest extractability for isoquercetin, and it was selected as the optimal solvent. Then, a series of mixtures of the tree leaves were prepared according to a simplex-centroid mixture design, and their DES-extracts were tested for skin-related activities, including antioxidant, anti-tyrosinase, and anti-elastase activities. The mixture design resulted in two special cubic models and one quadratic model best fitted for describing the antioxidant and anti-elastase activities, and the anti-tyrosinase activity, respectively. Based on the established models, three different optimal formulations of the three kinds of tree leaves were suggested for maximized responses. This strategy, based on the simplex-centroid mixture design with a DES as the extraction solvent, was proposed for the development of new materials from a mixture of natural resources, suitable for the cosmetics and related fields.

Finally, in their informative review, Detsi and Skarpalezos [6] attempted to summarize the use of deep eutectic solvents in the extraction of flavonoids, one of the most important classes of plant secondary metabolites. All of the applications reviewed reported success in isolation and extraction of the target compounds: competitive, if not superior, extraction rates compared with conventional solvents; and satisfactory behavior of the extract in the latter applications (such as direct analysis, synthesis, or catalysis).

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References

1. Loukri, A.; Tsitlakidou, P.; Goula, A.; Assimopoulou, A.; Kontogiannopoulos, K.; Mourtzinos, I. Green extracts from coffee pulp and their application in the development of innovative brews. *Appl. Sci.* **2020**, *10*, 6982. [CrossRef]
2. Dabetić, N.; Todorović, V.; Panić, M.; Redovniković, I.R.; Šobajić, S. Impact of deep eutectic solvents on extraction of polyphenols from grape seeds and skin. *Appl. Sci.* **2020**, *10*, 4830. [CrossRef]
3. Lakka, A.; Lalas, S.; Makris, D. Development of a low-temperature and high-performance green extraction process for the recovery of polyphenolic phytochemicals from waste potato peels using hydroxypropyl β-cyclodextrin. *Appl. Sci.* **2020**, *10*, 3611. [CrossRef]

4. Lakka, A.; Grigorakis, S.; Kaltsa, O.; Karageorgou, I.; Batra, G.; Bozinou, E.; Lalas, S.; Makris, D. The effect of ultrasonication pretreatment on the production of polyphenol-enriched extracts from *Moringa oleifera* L. (drumstick tree) using a novel bio-based deep eutectic solvent. *Appl. Sci.* **2020**, *10*, 220. [CrossRef]
5. Jin, Y.; Jung, D.; Li, K.; Park, K.; Ko, J.; Yang, M.; Lee, J. Application of deep eutectic solvents to prepare mixture extracts of three long-lived trees with maximized skin-related bioactivities. *Appl. Sci.* **2019**, *9*, 2581. [CrossRef]
6. Skarpalezos, D.; Detsi, A. Deep eutectic solvents as extraction media for valuable flavonoids from natural sources. *Appl. Sci.* **2019**, *9*, 4169. [CrossRef]