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Review Article

Green analytical chemistry in food safety: Eutectogel as an eco-friendly technology for the extraction and detection of pesticide residues in cereal grains

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Abstract

Global food security increasingly relies on effective analytical methods that monitor pesticide residues while minimizing environmental impact. Integrating green chemistry principles into food analysis is crucial for minimizing pollution, conserving resources, and enhancing food safety. This review explores eutectogels (Gelled Deep Eutectic Solvents) as sustainable materials for extracting and identifying pesticides in staple foods such as rice, wheat, and corn. By combining the solvation efficiency and environmental benefits of deep eutectic solvents (DES) with the practical advantages of solid-phase extraction materials, eutectogels offer a versatile platform for residue analysis. Recent developments have demonstrated their application in various microextraction techniques, including Solid Phase Micro-Extraction (SPME), Stir Bar Sorptive Extraction (SBSE), and Dispersive Micro Solid Phase Extraction (D-µ-SPE), achieving remarkable efficiency while significantly reducing organic solvent consumption. The customizable chemical composition of eutectogels enables targeted interactions with specific analytes and food matrices. This review critically evaluates current applications, assesses the greenness of eutectogel-based methods, and identifies key challenges, including reproducibility, commercialization, and regulatory acceptance, that must be addressed for widespread implementation.

Keywords: eutectogels, deep eutectic solvents, green analytical chemistry, pesticide residues, food safety

Introduction

The world population is projected to reach approximately 10 billion by 2050, underscoring the critical importance of food security [1]. For those focused on sustainable nutrition, it is important to note that rice, wheat, and corn together support nearly 60% of the global population [2]. However, intensive agricultural practices required to meet escalating food demands have led to increased pesticide use, raising concerns about residue levels in these vital crops. Recent monitoring studies reveal that more than 50% of agricultural samples, including fresh produce, contain pesticide residues, many of which exceed established safety limits [3].

To protect consumers and maintain consistency in food safety standards, global regulatory authorities have implemented Maximum Residue Limits (MRLs) [4]. The maximum residue limit (MRL) is the maximum allowed concentration of residue in a food product intended for human consumption. Nevertheless, the concentration of pesticide residue found in cereal grains varies significantly, influenced by factors such as the compound, origin, and processing stage. The limit of quantitation (LOQ) is defined as the lowest concentration of an analyte that can be reliably measured, typically fall within the µg/kg range [5]. For example, organophosphate pesticides such as Pirimiphos Methyl have been detected in wheat and maize at concentrations of 80–470 µg/kg [6]. In contrast, barley and rice often show elevated levels ranging from 10 to 1000 µg/kg, with approximately 14% of barley samples exceeding 1000 µg/kg [6].

The European Food Safety Authority (EFSA) considers validating the enforcement LOQ of 10 µg/kg for many cereal methods [4], while the Codex Alimentarius Commission has set maximum residue limits (MRLs) at 100 µg/kg for milled rice [7]. Meanwhile, the European Food Safety Authority (EFSA) has assessed an MRL of 50 µg/kg for the herbicide aminopyralid in maize, along with a limit of quantification (LOQ) of 10 µg/kg for the herbicide dichlorprop-P in cereal grains [8]. For analytical reliability, it is crucial to have LOQ below the MRL of

0.01 mg/kg, recovery rates between 70 and 120%, and relative standard deviation (RSD) not exceeding 20%. It is reported that eutectogel-based extraction methods have demonstrated limits of detection (LODs) in the range of 0.0003 to 6.6 μg/L and recoveries of 75 to 103%. These results are aligned well with these regulatory requirements, confirming their feasibility for compliance monitoring [6,7,8].

While conventional analytical methods are efficient, their reliance on hazardous chemical solvents and energy-consuming processes is inconsistent with the concepts of green analytical chemistry (GAC). GAC advocates incorporating the 12 principles of green chemistry into analytical methodologies, highlighting miniaturization of techniques, simplification of procedures, and the use of less hazardous reagents and solvents, with solvent-free extractions representing the most sustainable approach [9]. Conventional pesticide monitoring is slow, expensive, and ineffective. However, advancements such as the QuEChERS technique (Quick, Easy, Cheap, Effective, Rugged, and Safe) have improved accessibility and efficiency in this area [10].

In this context, Green Analytical Chemistry represents a transformative approach to sustainable analytical methods that reduce environmental impact while preserving analytical efficacy [11]. Deep eutectic solvents (DES) are an emerging class of green solvents, formed from simple, safe components, that combine the advantages of ionic liquids with lower cost and improved environmental compatibility, while offering unique properties such as low volatility, tunable polarity, and potential biodegradability [12]. Recent developments have led to the creation of eutectogels, which involve the immobilization of deep eutectic solvents (DES) within gel matrices. This innovative approach combines the solvation properties of DES with the mechanical strength of gels, demonstrating promise in energy storage, biomedicine, and environmental analysis [13].

Currently, research concentrates on DES-based microextraction techniques as effective, environmentally friendly mediums for analyzing pesticide residues in staple foods, such as milk. However, integrating eutectogels into these methodologies represents an advanced approach that combines solvent efficiency with the durability of solid-phase systems [14]. Gas chromatography (GC) and liquid chromatography (LC) combined with mass spectrometry are benchmark methods for pesticide detection, offering high sensitivity and selectivity for trace-level analysis [15].

Scope and objectives

This review aims to provide a thorough assessment of eutectogels as sustainable media for analyzing pesticide residues in rice and staple foods. The specific objectives include examining the fundamental principles of DES and eutectogel synthesis, evaluating current microextraction techniques using eutectogels, reviewing applications of eutectogel-based methods in the analysis of pesticides in staple foods, assessing the environmental sustainability of eutectogel-based methods, and identifying challenges and future perspectives in the field.

Deep eutectic solvents (DES) and their gelation

Deep eutectic solvents (DES) are composed of two parts: a hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD). DES exhibits a considerably lower freezing point compared to the individual components [16]. The most commonly utilized systems incorporate quaternary ammonium salts, such as choline chloride, as hydrogen bond acceptors (HBAs) and various organic compounds, including glycols, carboxylic acids, and sugars, as hydrogen bond donors (HBDs). These solvents have been proposed as environmentally friendly alternatives for synthesis processes. The unique combination of large, nonsymmetrical ions results in low melting temperatures due to reduced lattice energy [17].

Deep eutectic solvents (DES) share several favorable physical properties of conventional ionic liquids (ILs), such as low vapor pressure, non-flammability, and high tunability, offering significant practical and economic benefits. Compared with conventional ILs, their primary advantages include ease of preparation via simple mixing of inexpensive, readily available, and often well-characterized components. This not only lowers production costs but also facilitates large-scale application [17].

Natural deep eutectic solvents (NADES) are particularly noteworthy for their non-toxic, sustainable, and eco-friendly properties, making them potential candidates for a wide array of applications in biochemistry and green chemistry. Their distinctive combination of sustainability and utility positions them as innovative alternatives to hazardous synthetic ionic liquids currently in use, heralding a promising advancement in the field [18].

In an effort to replace hazardous organic solvents, cleaner techniques for identifying food toxins are being developed in alignment with the principles of green chemistry. Natural deep eutectic solvents (NADES) have emerged as a sustainable alternative. A recent study demonstrated the use of NADES-based dispersive liquid-liquid microextraction (DLLME) for the multi-residue analysis of pesticides in milk, followed by gas chromatography. This green analytical method achieved satisfactory analyte recoveries (64-89%) and low detection limits (0.9-3.9 ng/mL), validating NADES as a viable and efficient extraction medium [19].

The rationale for gelation: From DES to eutectogels

While DES offers numerous advantages as an extraction solvent, its liquid nature can limit its use in certain extraction methods, particularly those that require solid-phase handling. By transforming DES into semi-solid materials that retain the beneficial solvation properties of the liquid phase while enabling solid-phase extraction, gelation effectively addresses this limitation [20]. Gelation agents such as polymeric gelators (such as agarose, chitosan, and polyvinyl alcohol), low molecular weight gelators (such as fatty acids and amino acid derivatives), and inorganic gelators (such as clay minerals and silica nanoparticles). The resulting eutectogels provide mechanical stability and shape persistence necessary for extraction applications while preserving the microenvironment of the original DES [21].

Physicochemical structure and mechanisms of eutectogels

Eutectogels are characterized by their three-dimensional polymer network formed with deep eutectic solvents. These solvents facilitate the clustering of polymer chains via multiple noncovalent interactions, including hydrogen bonds, van der waals forces, polar interactions, and ionic interactions. In the polysaccharide-based eutectogel polymer, the network of hydrogen bonds formed by the DES is disrupted by the polymer, allowing for the formation of new hydrogen bonds with DES components. The stability of the gelation process is significantly influenced by these interactions. In other cases, altering the DES or water solution leads to crystallization of the polyvinyl alcohol polymer structure, forming a stronger, more homogeneous polymer network [22]. DES can also be polymerized or copolymerized to form polymeric DES (PDES), which can be encapsulated within an external gel matrix, such as calcium alginate beads, to produce a composite network with specific functional groups and sorption sites [23].

One of the principal mechanisms underlying adsorption is the hydrogen bonding that occurs between pesticide functional groups (e.g., nitro, carbonyl, heterocycles) and the hydrogen bond donor (HBD) and hydrogen bond acceptor (HBA) sites within the DES, a phenomenon observed frequently in molecular dynamics and quantum chemical studies. The ability of DES components to transfer charges to pesticides stabilizes the complexes formed between them, thereby enhancing extraction efficiency [22]. Electrostatic interactions, modulated by pH and ionic strength, play a critical role, as variations in the ionization state of pesticides and the acidic/basic conditions of the gel matrix affect electrostatic interactions, consequently influencing uptake. Additionally, hydrophobic partitioning has been observed in the DES-rich phase, with molecular dynamics studies demonstrating greater partitioning of pesticides in this phase than in aqueous solutions, attributed to van der waals and hydrophobic interactions. To augment surface adsorption, nanofillers such as magnetic Fe_3O_4/Fe_4O_4 nanoparticles or carboxylated multi-walled carbon nanotubes (MWCNT-COOH) have been introduced to increase the available surface area for adsorption [23]. The selectivity, capacity, and recyclability of eutectogels can be finely tuned through compositional design. The porosity and functional group density of the matrix are influenced by the choice of polymer and crosslinker, which, in turn, affect steric accessibility and the number of specific recognition sites for polar or nonpolar pesticides [22]. The composition of DES, HBD and HBA affects the hydrogen-bonding behavior, hydrophobicity, and charge-transfer properties of the matrix, thereby creating selective sorption sites.

Molecular dynamics and quantum chemistry simulations of different DES compositions revealed that varying DES composition alters distribution coefficients and selectivity toward certain pesticides, such as Acetamiprid and Imidacloprid [22]. By covalently integrating functional moieties during PDES production and copolymerization, customized pockets for target analytes can be created, leading to significant improvements in pesticide extraction effectiveness, up to 1.5-3.4 fold greater than that of conventional alginate beads when thymol methacrylic acid PDES is copolymerized with alginate [23]. The incorporation of magnetic particles and nanofillers not only enhances surface area and accessibility of specific adsorption sites but also improves the magnetic separation capacity, with enhancements ranging from 1.4 to 2.5 times when MWCNT-COOH and Fe_3O_4/Fe_4O_4 nanocrystals are utilized [23]. Moreover, water content and post-treatment can modulate polymer crystallinity and DES interactions, affecting mechanical recovery and long-term reuse. Solvent exchange or controlled water addition can strengthen polymer crystallinity and improve mechanical durability and resistance [22]. Several studies have shown that eutectogels are reusable for 9–40 cycles [22, 23, 24].

Synthesis and characterization of eutectogels

The synthesis of eutectogels typically follows in a two-step process: the formation of DES followed by gelation. The DES is first prepared by mixing the HBA and HBD components at an elevated temperature (typically 60-80 °C) until a homogeneous liquid is obtained. Following this, the gelatin agent is introduced either simultaneously with or after the formation of the DES, depending on the specific gelator used and the desired gel characteristics [25].

Table 1. Common DES compositions and their eutectogel applications in food analysis

HBA	HBD	Molar Ratio	Gelator	Application	Reference
Choline chloride	Glycerol	1:2	Agarose	Pesticide extraction from fruits	[26]
Choline chloride	Ethylene glycol	1:2	PVA	Mycotoxin analysis in cereals	[27]
Betaine	Lactic acid	1:2	Chitosan	Herbicide residues in rice	[28]
DL-Menthol	Decanoic acid	2:1	Silica gel	Organophosphorus pesticides	[29]
Choline chloride	Urea	1:2	Agar	Multi-residue analysis in wheat	[30]

Eutectogel-based microextraction techniques

Extraction strategies in analytical chemistry are transitioning from traditional liquid-liquid extraction (LLE) and solid-phase extraction (SPE) toward more effective and environmentally friendly alternatives [25]. Conventional methods often require large amounts of solvent, are time-consuming, and generate significant chemical waste. On the other hand, new analytical methods such as micro-dispersive solid-phase extraction (μ-dSPE) and magnetic solid-phase extraction (MSPE) require less solvent, are time-efficient, and yield higher recovery rates [31, 32]. The effectiveness of these analytical methods is highly determined by the adsorbent employed [33]. Eutectogels are multifunctional adsorbent gel matrices formed by immobilizing deep eutectic solvents (DES) in a three-dimensional polymer network [34]. These three-dimensional polymer networks provide stability, prevent leaching, and allow reusability. The advantages of DES include adjustable polarity and strong analytical affinity resulting from hydrogen bonding, pi-pi (π-π) interactions, electrostatics, and hydrophobic forces. Ongoing research is optimizing eutectogels for a variety of analytes across food, environmental, and biological samples [35, 36, 37]. Recent studies indicate that eutectogels are being used as analytical methods for their sensitivity, selectivity, and sustainability.

Eutectogel-based micro-dispersive solid-phase extraction (μ-dSPE)

Micro-dispersive solid-phase extraction (μ-dSPE) is a specialized type of dispersive solid-phase extraction (SPE) that uses micro-sized sorbents to enhance analyte interactions and reduce the use of harmful chemicals [38]. This method simplifies the analytical procedure by enabling direct dispersion within the sample [39,40]. The high surface-to-volume ratio in μ-dSPE improves mass transfer and recovery efficiency while reducing chemical waste [33,41]. However, limitations exist, including insufficient selectivity, structural instability, and susceptibility to leakage.

A study synthesized a cellulose-modified alginate coupled with a DES of adipic acid and L-cysteine for the extraction of phthalates from various matrices, including mineral water, apple juice, and peach juice. This approach used a porous eutectogel matrix with functional groups that promote hydrogen bonding and hydrophobic interactions, thereby achieving optimal extraction efficiency. The inclusion of cellulose bolstered the durability of the beads. For μ-dSPE, researchers utilized minimal amounts of beads dispersed in aqueous samples, followed by filtration, analyte desorption, and subsequent corona discharge ion mobility spectrometry (CD-IMS) analysis. This method demonstrated high analytical characteristics, with a LOD of 0.0003 μg L ^{1}, a linear range of 0.001 to 5.0 μg L^{-1}, recoveries of 91 to 103%, and RSD not exceeding 7%. The eutectogel beads exhibited remarkable reusability, maintaining consistent efficacy across more than forty extraction cycles. According to the AGREE assessment, the method described is considered environmentally friendly with a score of 0.73. The current research has proved that eutectogel-based μ-dSPE represents a sensitive, reproducible, and green approach for the detection of endocrine-disrupting phthalates in environmental water and food samples [42].

Eutectogel-based magnetic solid-phase extraction (MSPE)

Magnetic solid-phase extraction (MSPE) has gained popularity in sample preparation due to its magnetic properties, which allow for rapid and efficient separation of sorbents from sample matrices [43]. Compared to traditional SPE, MSPE (**Figure 1**) offers significant benefits, including faster extraction, reduced consumption of organic solvents, elimination of filtration and centrifugation, greater sorbent reuse, and more efficient separation [44, 45]. This method distinguishes itself by leveraging the dispersion of magnetic sorbents within the sample, followed by isolation via magnetic fields, facilitating a streamlined, effective workflow that enhances interactions between sorbents and analytes, ultimately improving mass transfer and overall extraction efficacy [44, 46, 47]. However, bare magnetic nanoparticles (Fe_3O_4) face challenges such as oxidation, agglomeration, and poor selectivity, which decrease their dispersibility and magnetic properties [48]. The modification of magnetic nanoparticles through the use of magnetic eutectogel offers potential solutions to these limitations, paving the way for more efficient extraction methodologies.

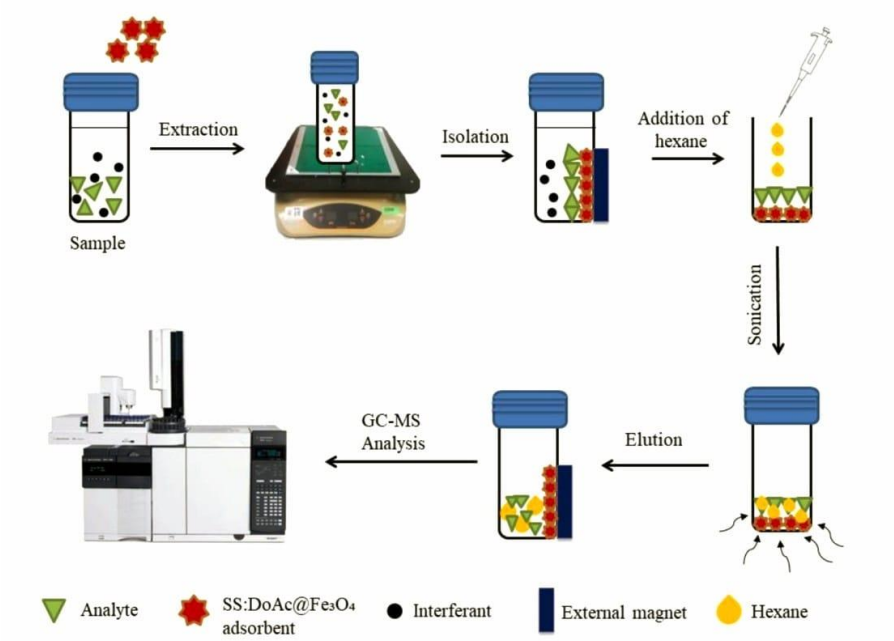


Figure 1. Graphical illustration of magnetic solid phase extraction (MSPE) procedure

In one study, a magnetic colloidal eutectogel comprising Fe_3O_4 XAD-16 nanocomposites and a choline chloride-phenol deep eutectic solvent was used as a sorbent for the extraction of carmoisine (E122) from candy, fruit juice, and jelly samples. This strategy involved dispersing the sorbent directly into the aqueous media, mixing to ensure uniform contact, magnetically separating the sorbent, and then desorbing the analyte for UV–Vis spectroscopy. The resulting method was proven highly effective, with a linearity range of 3 to 320 µg L^{-1}, R^2 of 0.9974, LOD of 0.86 µg L^{-1}, LOQ of 2.87 µg L^{-1}, and high reproducibility, as indicated by RSD < 9.2%. The application of eutectogel markedly enhanced the sorbent's selectivity and capacity, while its magnetic properties facilitated sorbent recovery. Therefore, the presented evidence confirmed that magnetic eutectogels can be considered a rapid, accurate, and inexpensive method for the determination of synthetic dyes in foods [49].

Recently, another study utilized Fe_3O_4 chitosan gel with DES to isolate fluoroquinolone antibiotics, namely ofloxacin and enrofloxacin, from milk. Herein, the magnetic eutectogel was mixed with the milk sample, allowing interaction between the analytes and the sorbent, and the analytes were subsequently isolated using an external magnet. The analytes were then desorbed with appropriate solvents and analyzed via HPLC-DAD. High sensitivity was observed, with LOD values of 0.0059-0.0067 µg mL^{-1} and LOQ values of 0.0198-0.0225 µg mL^{-1}. The recovery rate exceeded 91%, and the intra- and inter-day precision was lower than 8%. The linearity observed within the tested concentration ranges was confirmed, thereby meeting the established criteria for determining antibiotic residues in milk. This method eliminates the need for centrifugation or filtration, thereby streamlining the process and saving time. In addition, the researchers observe that the DES components derived from natural sources are less toxic than conventional organic solvents. These data confirm the efficiency of eutectogel-based MSPE in controlling the presence of veterinary drugs in milk, ensuring high precision and safety [50].

Application of pesticide analysis in staple foods

In analyzing pesticides in staple foods, it is essential to consider a range of chemical classes, including organochlorine, organophosphorus, carbamate, pyrethroid, triazine, and neonicotinoid compounds. Each of these classes presents unique analytical issues related to their polarity, volatility, and stability at elevated temperatures [39]. Contemporary approaches to pesticide analysis use chromatography coupled with mass spectrometry for identification and quantification. Gas chromatography-tandem mass spectrometry (GC-MS/MS) is employed to analyze volatile and stable pesticides, such as organochlorine and pyrethroid pesticides. Meanwhile, liquid chromatography-tandem mass spectrometry (LC-MS/MS) is used to analyze polar and thermally labile pesticides, such as carbamates and neonicotinoids. Recent validation experiments have demonstrated that modern LC-MS/MS and GC-MS/MS instruments can simultaneously analyze over 400 pesticides, with detection limits well below regulatory levels [43]. Furthermore, the combination of eutectogel extractions with these technologies promises significant advances in terms of sensitivity and matrix suppression.

Critical review of applications in rice and other staples

Rice analysis

The analysis of rice presents challenges due to its complex matrix composition, which includes elevated starch levels and various interfering substances that may affect extraction efficiency and lead to matrix effects [40]. Recent research has demonstrated the successful application of DES-based extraction methods for the analysis of pesticides in rice samples. One study applied a modified QuEChERS approach combined with GC-MS/MS technique for simultaneous extraction and analysis of four different pesticide residues, achieving recovery rates of 70 to120% and a relative standard deviation less than 20% [44]. Although neither DES nor eutectogels were used in this study, the potential of DES-modified eutectogels is clearly demonstrated. Low transition temperature mixtures (LTTMs) have potential for analyzing rice samples. This system exhibits characteristics similar to those of DES regarding environmental friendliness and extraction efficiency. It is distinguished by its tunable polarity, which facilitates the selective extraction of different pesticides from rice.

Wheat and corn analysis

Wheat and corn differ from rice in lipid concentrations and the presence of interfering proteins during extraction [45]. The use of the eutectogel technique in these matrices has been successful, especially for the extraction of polar pesticides that are challenging to extract using traditional methods. The electrospun nanofibrous eutectogel material has been shown to be effective for the extraction of multi-class pesticides from cereal samples [46].

Cross-matrix validation

The availability of universal extraction techniques based on eutectogels, adaptable to various types of staple foods, is a significant strength for analysis. Research findings indicated that when appropriate DESs are used, the extraction technique becomes reliable regardless of the food matrix type [47].

Greenness assessment

As attention to the environmental effects of analytical methods has increased, several indices have been developed to quantify the "greenness" of analytical methodologies. Notable among these is the green analytical procedure index (GAPI), which evaluates all aspects of the procedure, starting from sample extraction through waste production [48]. Additionally, the qualitative Analytical Eco-Scale, which quantifies penalty scores for the use of hazardous compounds, with lower scores indicating a more environmentally friendly analysis [49]. Furthermore, the recently developed Analytical GREEnness Metric (AGREE) adds to this framework [50].

Table 2 clearly demonstrates the superior environmental profile of eutectogel-based methods compared to conventional approaches. The dramatic reduction in organic solvent consumption, combined with lower energy requirements and reduced waste generation, positions these methods as truly green alternatives for pesticide residue analysis.

Greenness scoring criteria and mechanistic insights on green metrics

To thoroughly understand the greenness analysis presented in Table 2, it is important to describe the scoring criteria and mechanistic insights for each metric, and to demonstrate how the eutectogel adsorbent improves the greenness profile of the extraction method.

Eco-scale scoring and interpretation

The Eco-Scale method starts with an initial score of 100 and subtracts penalty points for various non-green aspects of the analytical procedure. Points are deducted for the use of hazardous reagents, large solvent volumes, high energy consumption, and waste generation [54]. The final eco-score is obtained by subtracting the total penalty points from 100, with higher scores indicating more environmentally friendly practices. Eco-scores of 75 or above are

Table 2. GREEnness assessment of extraction methods for pesticide analysis [51,52,53]

Method	Organic Solvent (mL/sample)	Energy Consumption	Waste Generation	GAPI Score	Eco-Scale Score*
Conventional QuEChERS	10 - 15	Moderate	High	45	65
DES-assisted extraction	2 - 5	Low	Low	75	85
Eutectogel D-μ-SPE	1 - 3	Low	Low	80	88
Eutectogel-SBSE	0.5 - 2	Low	Low	85	90
Eutectogel-SPME	0.1 - 0.5	Very Low	Very Low	90	95

considered excellent in the context of green chemistry; scores between 50 and 75 are acceptable; and scores below 50 are considered poor. Published methods employing eutectogel have demonstrated eco-scores ranging from 88–95, attributed to minimal penalty points from reduced solvent use, lower toxicity of DES components, and reduced waste [54].

AGREE scoring and interpretation

The AGREE (analytical greenness metric) method assesses all 12 principles of green analytical chemistry to a 0 to 1 scale, resulting in a circular pictogram with 12 wedges, each representing one principle [55]. This scoring method aggregates scores for all 12 principles (with user-defined weights) and produces a final composite score ranging from 0 to 1. Scores closer to 1 indicate greater compliance with the 12 green analytical chemistry principles. The pictogram visually highlights the principles (reagent amounts, waste, energy, miniaturization), which may require further improvement. In a specific experiment, a eutectogel-based dispersive micro-solid-phase extraction (D-μ-SPE) achieved an AGREE score of 0.73, indicating good overall greenness [55].

GAPI score and interpretation

The green analytical procedure index (GAPI) evaluates the overall environmental sustainability of a comprehensive analytical method, covering all phases from sample collection to final determination. GAPI employs a distinctive five-pentagram symbol to gauge the environmental impact of each step in the analytical process, with color coding: green for low impact, yellow for medium impact, and red for high impact. GAPI's qualitative and descriptive nature often complements numeric metrics such as Eco-Scale and AGREE, providing a more comprehensive view of environmental sustainability [56].

Red, green, blue (RGB) and white analytical chemistry (WAC)

The RGB or WAC method integrates red values related to functionality, green values reflecting environmental impact, and blue values emphasizing practicality, to generate whiteness values [56]. Each parameter is computed separately, providing an integrated view of the technique's effectiveness. Higher or "whiter" values suggest a successful balance between greenness and functionality while maintaining performance efficiency [56].

Evaluation of overall procedure vs. sorbent contribution

It is important to clarify that the values shown in the greenness assessment in **Table 2** relate to the entire extraction process, not just the performance of the eutectogel alone. The eutectogel sorbent significantly enhances the sustainability of the extraction procedure through several key factors. Notably, it drastically reduces solvent requirements, with eutectogel-based techniques requiring only 0.1-3 mL of solvent per sample, compared to 10-15 mL for traditional QuEChERS techniques. Additionally, this method offers high reusability, as demonstrated by its effectiveness in over 40 extractions across various studies, thereby decreasing the solid waste generated per sample. Also, this method uses biodegradable, nontoxic ingredients such as choline chloride, organic acids, and natural hydrogen bond donors, thereby decreasing the toxicity score in the Eco Scale and improving AGREE principles regarding reagent use. Finally, because sorbents can be reused, they generate less solid waste per sample than one-time-use alternatives [54,55,56].

Challenges and future perspectives

Challenges: Reproducibility

A significant challenge associated with eutectogel-based extraction is ensuring consistent reproducibility across laboratories and among operators. The synthesis of eutectogels is highly dependent on specific conditions, such as the type and molar ratio of DES components, gelator concentration, preparation temperature, and mixing conditions. Each of these factors plays a crucial role in determining the final physicochemical properties and extraction efficiency [57]. The inconsistency in the quality of raw material, particularly those derived from natural-based components, further complicates standardization. Additionally, eutectogels are often sensitive to moisture content and storage conditions, while temperature-dependent gelation behaviors can lead to variability between batches [58].

Experimental evidence confirms this variability. For instance, a study reported that water content significantly affects the hardness, adhesiveness, viscoelasticity, and thermosensitivity of choline chloride-xylitol eutectogels [59]. Furthermore, another study found that durian seed gum concentration and the composition of NADES resulted in differences in viscosity, yield stress, and stability, thereby affecting performance [60]. Also, in another study, eutectogels produced through carefully monitored heating-cooling processes exhibited constant rheological parameters even at high temperatures, underscoring the importance of controlling preparation conditions to obtain reproducible results [61]. Even slight variations in composition and preparation conditions can lead to inconsistent results. Therefore, there is a need to standardize production protocols and quality standards, and to incorporate inter-laboratory validation trials to evaluate extraction performance.

Commercialization

Scaling up eutectogel-based methods from bench research to commercial applications presents multiple

barriers. Laboratory syntheses often rely on small-scale, carefully controlled conditions that are difficult to replicate in industrial processes. During scale-up, variations in mixing efficiency, heat transfer, and component purity can lead to heterogeneity in gel strength, porosity, or extraction performance [62]. Achieving inter-batch consistency is particularly problematic due to the sensitivity of eutectogels' properties to even minor differences in the composition of DES, gelator purity, and water content [56].

These problems are highlighted by empirical evidence. For instance, a study reported that increasing the concentration of durian seed gum in the formulation enhanced its barrier properties and viscoelasticity. However, this improvement also led to higher moisture absorption capacity and structural weaknesses in certain formulations, suggesting that large-scale production would lead to performance variations [60] Furthermore, obtaining regulatory clearance poses another challenge, as most DES used for gelation lack sufficient safety data for direct food contact. Additionally, cost is a challenge, as sourcing, purifying, and formulating NADES and eutectogels can be more expensive than conventional extraction methods such as QuEChERS and SPE [62]. Lastly, there are challenges concerning intellectual property, as many of the formulations used are unique and not available in open-source databases.

Comprehensive toxicity assessment

Although eutectogels are often promoted as green alternatives to traditional solvents, there is a notable lack of comprehensive toxicity and safety information. While DES components are generally recognized as safe, their toxicity depends strongly on the specific characteristics and proportions of hydrogen bond donors (HBDs) and hydrogen bond acceptors (HBAs) involved [62]. Some sugar-based or alcohol-based DES have low cytotoxicity, whereas organic acid-based DES may exhibit significant cytotoxicity even at low concentrations [56]. Eutectogels incorporate gelators and structural modifiers, potentially altering their overall toxicological behavior [58].

Recent research has made preliminary observations. The cytotoxicity of choline chloride-xylitol eutectogels was lower in vitro than that of hydrogels; however, this assessment was conducted over a short-term period. Other comprehensive studies encompassing chronic and genotoxicity testing, as well as in vivo tests, are currently lacking. There are also environmental implications of these gels that warrant consideration, even though some are biodegradable. The life cycle assessments for their production and disposal are not yet available [62].

Method automation

Integrating eutectogel-based methods into automated laboratory workflows presents practical challenges due to their semi-solid, viscous nature. High viscosity complicates accurate pipetting and dispensing, as standard liquid-handling systems lose precision with viscous materials [63]. Automated dispensing of eutectogels will require specialized valves and temperature control to achieve reproducibility. Similarly, temperature fluctuations influence the rheological stability of eutectogels, as shown in studies on choline chloride–xylitol systems [51]. The identified factors indicate that implementing specialized automation strategies, such as enhanced dispensing tools and rigorous temperature control, will be essential before integrating eutectogel-based extraction into high-throughput or regulatory laboratory setting.

Future perspectives: Advanced materials development

Future research should aim to develop eutectogel materials with enhanced selectivity, operational stability, and ease of handling. Although molecularly imprinted eutectogels remain unexplored, the successful use of molecular imprinting in DES-based polymers suggests a potential avenue for future research [64]. DES-assisted Molecularly Imprinted Polymer (MIP) systems have demonstrated strong binding specificity in other contexts, suggesting that combining imprinting with eutectogel matrices could be a viable approach [65].

Magnetic eutectogels represent a second promising direction. Several studies have shown that magnetic nanoparticles, such as Fe_3O_4 embedded in DES-based gels or micro-eutectogels, enable rapid magnetic separation and simple extraction workflows in food matrices. This includes validated applications for melamine in milk and for antibiotic residues in foodstuffs [66, 67]. These findings, presented in these works, demonstrate the feasibility and the analytical performance gains achievable with magnetic eutectogel designs.

Stimuli-responsive eutectogels are another promising concept. DES-enhanced smart hydrogels exhibited tunable responses to pH, temperature, and other triggers, potentially enabling controlled extraction and release in eutectogel systems [68]. Finally, advances in the design of hierarchical porous materials could further improve eutectogel performance. Research on porous DES-based hydrogels and hybrid electrolytes indicated that introducing multi-scale porosity enhances surface area and mass transfer properties [69]. Applying this concept to eutectogels would help overcome diffusion limitations and accelerate analyte uptake, enabling more efficient extraction in time-sensitive workflows.

Expanded applications

Although pesticide monitoring has been the primary

focus, eutectogel platforms are adaptable to a broader range of food safety challenges. The presence of mycotoxins in cereals, veterinary drug residues in animal-derived foods, and additive contaminants signifies a rational progression for future applications. Furthermore, eutectogels can support nutritional profiling (e.g., vitamins, amino acids) and authenticity testing. This capability enhances the versatility established through DES-based extractions in food matrices [70]. The expansion of eutectogels will play a crucial role in establishing their value, not only in the realm of pesticide regulation but also as versatile analytical instruments in ensuring food and environmental safety.

Method validation and standardization

The effective implementation of analytical methods requires comprehensive validation in accordance with established international guidelines, such as those set by the International Council for Harmonisation (ICH), the U.S. Food and Drug Administration (FDA), and the European Medicines Agency (EMA). These guidelines emphasize the importance of laboratory-scale validation, inter-laboratory comparisons, proficiency testing, and the use of certified reference materials (CRMs). The alignment of protocols is essential to ensure comparability between laboratories and facilitate the transition of new methodologies from proof-of-concept studies to routine analytical practice.

Regulatory acceptance

To achieve comprehensive regulatory acceptance, eutectogel-based methods must demonstrate analytical equivalence with established techniques while offering clear advantages in safety, sustainability, and cost. Regulatory bodies will expect robust validation packages supported by toxicological and environmental assessments. Cost–benefit analyses will be equally important, especially in official food control laboratories, where operational efficiency is critical. Demonstrating these attributes will place eutectogels as a viable, green alternative to conventional extraction methods.

Conclusion

Eutectogels represent a paradigm shift toward sustainable analytical chemistry in food safety applications, offering a unique combination of green chemistry principles and analytical performance. By synergistically combining deep eutectic solvents with gel matrices, they create adaptable extraction platforms that substantially reduce the environmental impact associated with traditional extraction methods while addressing many of those methods' inherent limitations.

Despite the promising capabilities of eutectogels, it is essential to highlight the existing gaps in the literature regarding their application for pesticide extraction from cereal-based samples. The field has not yet progressed to the proof-of-concept stage for cereal matrices, and there is a lack of comprehensive research evaluating the efficacy of this extraction technique specifically for cereals. To establish its applicability across various cereal crops—such as wheat, rice, corn, and barley—further empirical studies are warranted. The LOD obtained through eutectogel extraction, ranging within the limits of low µg/kg and recoveries of 75 to 103%, complies with MRL regulations, which are equal to 10 to 100 µg/kg. However, validation studies are needed to implement these methods in laboratory practice. Recent literature suggests a growing interest in the application of eutectogels for analyzing pesticide residues in staple foods, with findings indicating remarkable extraction efficiency across diverse pesticide classes, particularly for polar compounds that are challenging to extract with conventional methods. Recent literature demonstrates the growing interest and promising results in eutectogel applications for pesticide residue analysis in staple foods. However, these materials show excellent extraction efficiency across diverse pesticide classes, with particular advantages for polar compounds that are challenging for conventional methods of extraction. However, several challenges must be addressed before widespread implementation can be achieved, including regulatory acceptance, which requires the establishment of standardized synthesis protocols, robust quality control measures, and clearly defined validation criteria.

Key areas requiring targeted research include resolving reproducibility concerns, overcoming commercialization barriers, conducting thorough toxicity assessments, and facilitating process automation. Future research should also focus on advanced material development, expanded applications, comprehensive method validation, and regulatory collaboration. Innovations such as molecularly imprinted, magnetic, and stimuli-responsive eutectogels demonstrate substantial potential for the emergence of next-generation extraction materials. Additionally, the adaptability of these materials can be further evidenced by expanding their applications beyond pesticide analysis to include mycotoxins, veterinary drugs, and other food contaminants. From a practical standpoint, the success of eutectogel-based methods ultimately depends on their ability to meet the stringent requirements of regulatory agencies while providing clear advantages over existing approaches. By integrating environmental sustainability, analytical performance, and economic viability, eutectogels position themselves as promising candidates for the future of green analytical chemistry in food safety. The adoption of sustainable analytical methods underscores the importance of these innovations, which not only address current analytical challenges but also uphold the analytical rigor required for food

safety assurance. Achieving the full potential of these advanced technologies to fortify global food security requires persistent research and development efforts, collaboration with industry stakeholders, and active involvement from regulatory bodies.

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