



Recent Advances in Molecular-Imprinting-Based Solid-Phase Extraction of Antibiotics Residues Coupled With Chromatographic Analysis

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Abuse and residues of antibiotics cause great harm to organisms and the environment. Appropriate sample pretreatment is usually required for sensitive determination, because of the low content presence of a variety of antibiotics in complicated matrices. Molecular-imprinting-based solid-phase extraction (MISPE) has been widely used for sample pretreatment of antibiotics, using molecularly imprinted polymers (MIPs) as adsorbents. Herein, we comprehensively review the recent advances of MISPE of antibiotics, followed by chromatographic analysis. Various solid-phase extraction (SPE) modes based on MIPs are briefly introduced, such as conventional SPE, dispersive SPE, magnetic SPE, matrix solid-phase dispersion, and pipette-tip SPE. Then, several emerging preparation techniques for antibiotics MIPs are summarized including surface imprinting, nanoimprinting, living/controlled radical polymerization, multitemplate imprinting, multifunctional monomer imprinting, and dummy template imprinting. Subsequently, applications of MISPE to analysis of a variety of antibiotics residues since 2018 are overviewed, including sulfonamides, quinolones, tetracycline, and others. Finally, the preparation and application of antibiotics MIPs are prospected.

Keywords: molecularly imprinted polymers, antibiotics, sample preparation, solid-phase extraction, review, imprinting technique

INTRODUCTION

Antibiotics are produced by bacteria, molds, or other microorganisms in the course of life, which can interfere with or inhibit the survival of pathogenic microorganisms (Fleming, 1929; Stachelek et al., 2021). They can be categorized into seven main kinds, namely, sulfonamides (SAs), quinolones (QNs), tetracyclines (TCs), macrolides (MALs), chloramphenicol (CAPs), β -lactams (BLAs), and aminoglycosides (AGs) (Chen et al., 2017; Ming et al., 2021). Besides being widely used to prevent

and treat animal and human diseases, antibiotics are also widely used as growth-promoting agents in animal husbandry and aquaculture, playing an important role in improving animal and human health (Ming et al., 2021). However, with the increasing use and abuse of antibiotics, bacteria are quickly adapted to the antibiotics, and all kinds of “superbugs” are being born (Stachelek et al., 2021). Consequently, antibiotic residue has become one of the most important environmental issues in the world. The residues and accumulation through various pathways in animals and the environment not only induce the growth of drug-resistant bacteria with increasing numbers and species (Stachelek et al., 2021) but also accumulate toxicity through the food chain, causing great harm to the ecological environment and human health (Liu et al., 2020). It is urgently required to develop effective analytical methods toward antibiotics residues. Chromatography and mass spectrometry (MS) based methods are commonly utilized, especially high-performance liquid-phase chromatography- (HPLC-) and tandem MS (HPLC-MS/MS). Despite high sensitive detection techniques, such as HPLC-MS/MS, it is still quite imperative to use influential sample preparation/pretreatment steps prior to detection, owing to the characteristics of low residual amounts, various interfering factors, and complicated sample matrices (Dugheri et al., 2021).

At present, sample pretreatment is widely employed in the analysis of trace analytes in complex matrices to purify and enrich the analytes and thereby will improve the sensitivity and accuracy of analytical methods (Zhang Y. et al., 2020). Currently, solid-phase extraction (SPE) that uses solid adsorbent to adsorb the target analytes is commonly used in the process of sample pretreatment, since it has many advantages, such as less time-consuming, simple operation, high efficiency, low or without solvent, and good compatibility with different analytical methods (Wu et al., 2021). SPE has a variety of modes, mainly including conventional SPE, dispersive SPE (DSPE), magnetic SPE (MSPE), matrix solid-phase dispersion (MSPD), solid-phase microextraction (SPME), stir-bar sorptive extraction (SBSE), and pipette-tip SPE (PT-SPE). It is well known that solid adsorbent is the key parameter of SPE efficiency. Nowadays, commercially available solid adsorbents like HLB, C18, and Oasis MCX usually exhibit nonspecific adsorption on the target analytes, which decrease the selectivity and specificity for antibiotics, thus greatly limiting the application of trace analysis of antibiotics in complicated matrices. Molecularly imprinted polymers (MIPs) with structure predictability, recognition specificity and application universality, easy preparation, low cost, and so on have gained great popularity as SPE adsorbents (Li et al., 2013; Chen et al., 2016; BelBruno, 2019; Arabi et al., 2020).

MIPs are prepared by molecular-imprinting technology based on the principle of antigens and antibodies specifically recognizing and binding, and the technology has often been vividly described as the “artificial lock” technology for preparing to recognize “molecular key” (Li et al., 2013). MIPs have been widely used for sample pretreatment (Płotka-Wasyłka et al., 2016), chromatographic separation (Wang and Cao, 2015; BelBruno, 2019), chemical/biosensors (Chen et al., 2016; Gaudin,

2020; Arabi et al., 2021), and other fields (BelBruno, 2019; Haupt et al., 2020). Using MIPs as the adsorbents in SPE, molecularly imprinted or molecular-imprinting-based SPE (MISPE) can specifically recognize targets, which can effectively adsorb targets in complex matrices. Therefore, MISPE is extensively used for the highly selective cleanup and enrichment of trace antibiotics in complex samples. Furthermore, MISPE combined with separation and detection technologies such as chromatography has been swiftly developed and applied to achieve rapid, simultaneous, selective enrichment, and sensitive detection of multiple antibiotics contaminants (Li and Row, 2017; Wang J. et al., 2018; Wang S. et al., 2018; Hu et al., 2018; Zhu et al., 2019b; Lu et al., 2019).

Therefore, in this work, the application of MISPE coupled with chromatographic analysis of antibiotics is reviewed comprehensively, focusing on the recent advances since 2018. Firstly, the classification of SPE and the commonly used advanced preparation techniques of MIPs are overviewed. Then, the applications of MISPE for the analysis of a variety of antibiotics are emphasized. Finally, some attempts to facilitate the wide application of MISPE in the field of sample pretreatment are proposed.

VARIOUS SPE MODES BASED ON MIPS

SPE is a technology to separate the component to be tested from the interfering component by the difference of the adsorption ability of solid adsorbent on each component in the liquid sample. The separation efficiency is increasing with the improvement of the selectivity of solid adsorbent toward target analyte (Fan, 2020). Therefore, the use of highly selective MIPs as SPE adsorbent can achieve efficient extraction and enrichment of target analytes, which has been widely used in antibiotics residues detection (Lian and Wang, 2018; Xie et al., 2018; Hammam et al., 2019; Negarian et al., 2019; Zhu et al., 2019b; Tan et al., 2020; Yu et al., 2020). Various SPE modes based on MIPs, i.e., conventional SPE, DSPE, MSPE, MSPD, SPME, SBSE, and PT-SPE, are briefly introduced below.

SPE generally means the conventional SPE, namely, packed SPE, and it is a common method for sample preparation. The column is packed with a solid adsorbent to make the sample be tested flow through the column so that the solid adsorbent can adsorb the target compounds, and then the target compounds can be separated and enriched by chemical reagent elution or heating desorption. The high selectivity of MIPs can greatly improve the extraction efficiency of SPE. The SPE with MIPs as extraction adsorbent has been widely used in the determination of antibiotics (Huang L. et al., 2019; Ma and Row, 2019; Qin et al., 2020).

DSPE does not need the packing and washing steps, the extraction time is short, and the adsorbent can be more fully dispersed into the sample solution to improve the adsorption effect. After the purified sample is shaken and centrifuged, the supernatant can be directly or simply processed into the next step of the analysis. This method proves to be quick, easy, cheap, effective, rugged, and safe and is also known as the QuEChERS method. As an efficient and rapid sample pretreatment

technology, DSPE is widely used in the analysis and detection of contaminants and antibiotics residue analysis (Song et al., 2018; Lu et al., 2019).

MSPE is a technique in which a magnetic or magnetizable material is used as a solid adsorbent for SPE. Instead of being packed in the extraction column, a magnetic adsorbent is directly added to the sample solution or suspension so as to attain a full dispersion. The target analyte is adsorbed on the surface of the dispersed magnetic adsorbent, and an external magnetic field is utilized to separate the target analyte from the sample matrix (Liang J. et al., 2018). MSPE requires only low consumption of adsorbents and equilibrium time to realize the enrichment and separation of trace analytes. MSPE can avoid column blockage, which is a very important problem in traditional SPE (Li et al., 2019). Magnetic MIPs (MMIPs) materials have been widely used in the detection of many types of antibiotics (Liu et al., 2017; Nazario et al., 2017; Kunsu-Ngiem et al., 2018; Li Z. et al., 2018; Qin et al., 2019; Dil et al., 2021; Gao et al., 2021; Lamaoui et al., 2021).

MSPD is the basic process as follows: adding solid-phase adsorption materials directly to the sample matrix, mechanical mixing resulting in a semidry mixture, using the obtained mixture as a packed column, cleaning the column with a small amount of reagent to remove impurities, and finally eluting the target analyte by a small amount of eluent. MSPD combines with sample crushing, extraction and purification, which not only avoids sample loss but also save solvent/time. MSPD has the advantages of simple and quick operation, high extraction efficiency, no special equipment, and so on; large quantities of automatic analysis can be carried out through MSPD. Therefore, MSPD is widely used in the analysis of antibiotics residues (Wang S. et al., 2018; Soares et al., 2021), contaminants (Liang et al., 2019), and harmful components (Tang et al., 2019). As a kind of solid-phase adsorbent with high selectivity, MIP has been used in MSPD, which provides powerful technical support for the analysis of pollutants (Wang S. et al., 2018) and harmful components (Balsebre et al., 2018) in a complex matrix.

SPME is a solvent-free pretreatment method developed on the basis of SPE. It is easy to be combined with gas chromatography (GC), HPLC, and capillary electrophoresis (CE), so it has been widely used (Barahona et al., 2019; Huang S. et al., 2019; Guo et al., 2020). SPME has been successfully applied in the analysis of organic and inorganic substances in gas, water, soil, sediment, and other environmental samples (Reyes-Garces et al., 2018). SPME process is actually the adsorption/desorption process of each component in the sample on the surface of the extracted fiber coating, and its principle depends on the distribution balance between the analyte in the sample matrix and the extraction phase. By selecting different groups of coating heads, SPME can attain ideal extraction efficiency toward targeted components, while other components are restrained. The MIP coating, because of its simple preparation, good repeatability, high mechanical strength, good resistance to high temperature, and solvent resistance, especially the advantage of efficient choice specificity for trace target in complex environment medium analysis, has a broad application prospect and become a research hotspot of selective SPME coatings (Barahona et al., 2019; Liu et al., 2019).

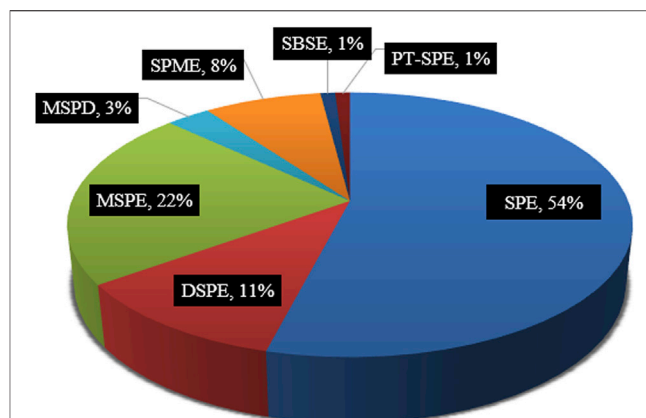


FIGURE 1 | Relative occurrence of the various samples pretreatment methods combined with MIPs detection antibiotics residual form 2018 to 2021 in this review. This data was extracted from Web of Science on June 08, 2021. [TS = imprint * AND solid phase extraction (or dispersive solid phase extraction, magnetic solid phase extraction, matrix solid phase dispersion, pipette-tip solid phase extraction, respectively)* AND antibiotics] NOT AU-Smalley RE.

SBSE is a variant of SPME, in which the glass tube with an inner magnetic core is coated with an extractive adsorption coating. After the distribution balance between the sample matrix and the coating on the surface of the stirring rod is reached, the sample is analyzed by thermal desorption or solvent desorption technology. Polydimethylsilane (PDMS) is the widely used coating material of SBSE. The coating prepared by the sol-gel method is compact in structure, highly hydrophobic, and stable in chemical properties. It is suitable for the extraction of nonpolar and weak polar compounds in the water phase. The SBSE has been developed to improve the sensitivity by using a larger volume of extraction phase. The difference is that a magnetic core is required for self-stirring to accelerate mass transfer, as thicker extractive coatings are usually prepared. However, since the SBSE format with magnetic agitation is not as convenient for sampling *in vivo* or in the field as fiber and film shapes, SBSE is commonly used for in-bottle extraction (Rodriguez-Gomez et al., 2018). However, the MIP coating-based SBSE is rarely used in antibiotic detection (Cui et al., 2021; Yang et al., 2017).

PT-SPE fills the pipette head with an adsorbent, making SPE setup smaller and analysis more environmentally friendly. PT-SPE is currently the most concerned SPE modes, mainly because it requires fewer adsorption materials, significantly reduces organic solvents consumption, and saves the cost. This method is simple and does not need extra special instruments. The transfer and distribution mechanism of the analytes to be enriched is roughly the same as that of the traditional SPE. The eluted solution can be directly used for LC or MS analysis without vacuum concentration. The pipetting nozzle can realize sampling, purification, enrichment, and quantification, at the same time, which makes up for the technical defects of traditional pretreatment methods, such as complicated operation, large amount of organic solvent, and easy loss of targets (de Oliveira et al., 2016; Teixeira et al., 2018; Zhang Y. et al., 2020; Hashemi et al., 2019).

Relative occurrence of the above-mentioned MISPE toward antibiotics residues detection within 2018–2021 is shown in

TABLE 1 | The main characteristics, advantages, and disadvantages of SPE.






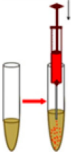

SPE mode	Schematic	Main characteristics	Advantage	Disadvantage
SPE		No phase separation operation; easy to realize automatic batch processing.	Simplicity and flexibility; influential clean up; having high enrichment factor; high recovery rate and wide application.	High consumption of organic solvents and sorbent; incompatible with solid sample; channeling or blockage of cartridge.
DSPE		No need to wash; short extraction time; the adsorbent can be more fully dispersed into the sample solution to improve the adsorption effect.	Higher efficiency and faster compared with packed SPE; no need of conditioning; short extraction consumption of organic solvents.	Possibility of spoiling the sorbent in complex sample; incompatible with solid sample; remaining residual of sorbent sample solution.
MSPE		Magnetic and magnetically modified adsorbents have high adsorption extraction capacity.	Simplicity of operator; high selectivity; short extraction time and low cost; green; large specific surface area; superparamagnetism; multiple adsorption sites.	Magnetic nanoparticles (Fe_3O_4) have poor selectivity and affinity for oxidation, aggregation, and adsorption; fewer varieties available; some materials are magnetically unstable; the practical application device is not perfect and the degree of automation is low.
MSPD		Extraction, filtration, and purification can be completed in one step without the need for tissue homogenization, precipitation, centrifugation, pH adjustment, and sample transfer, thus avoiding the loss of samples.	Simple usage; cost effectiveness; compatible with all liquid, semisolid, and solid samples.	High backpressure or blocking the cartridge; inability to reuse the sorbent.
SPME		Sampling and enrichment are carried out simultaneously; combined with GC, sampling, enrichment, and injection can be achieved in one step to reduce sample loss; matrix consumption can be ignored; suitable for field sampling and analysis.	Compatible with liquid, gaseous, and solid samples; solvent-free technique; clean extraction; ease of automation.	High cost; lack of robustness; bending the needle; breaking the fiber; flawed extraction.
SBSE		Sample is analyzed by thermal desorption or solvent desorption technology; no external agitator is needed, competitive adsorption can be avoided, and extraction enrichment can be realized while stirring itself.	Simplicity; high preconcentration capacity.	Time-consuming and error-prone manual extraction steps; high solvent consumption.
PT-SPE		By filling the pipette head with an adsorbent, the eluted solution can be directly used for LC or MS analysis without vacuum concentration. The pipetting nozzle simultaneously completes sampling, quantification, purification, and enrichment, which makes up for the technical defects of traditional pretreatment methods such as complicated operation, large amount of organic solvent, and easy loss of target material.	Low consumption of sample, sorbent, and organic solvents; cheapness; accessible tools; simplicity of handling; fast automation.	Blocking pipe-tip; leakage of sorbent; incompatible with solid samples.

Figure 1. As seen, conventional SPE is the most frequently used (54%), followed by MSPE and DSPE at 22 and 11%, respectively, and the two modes of SPME and MSPD are not higher than 10%, while PT-SPE and SBSE are both just 1%. The main characteristics, advantages, and disadvantages of the seven types of SPE techniques are listed in **Table 1**, with a reasonable expansion and modification of **Figure 1** of our previous work (Arabi et al., 2020).

EMERGING TECHNIQUES FOR MIPs PREPARATION

Various efficient MIPs toward antibiotics have been prepared by virtue of emerging techniques such as surface imprinting,

nanoimprinting, and living/controlled radical polymerization (LCRP) technology and multitemplate, multifunctional monomer, and dummy template imprinting strategy. **Figure 2** schematically illustrates the six techniques' basic processes/mechanisms.

Surface Imprinting

Surface imprinting technology means preparing imprinted materials by controlling templates to locate at the surface or in the proximity of materials surface to create more effective recognition sites (Chen et al., 2016). It can overcome the disadvantages of low binding capacity and difficulty in elution of traditional MIPs. Core-shell structured MIPs are the major type of surface imprinting MIPs, owing to the increased surface area and larger binding capacity, and thus, they are widely used

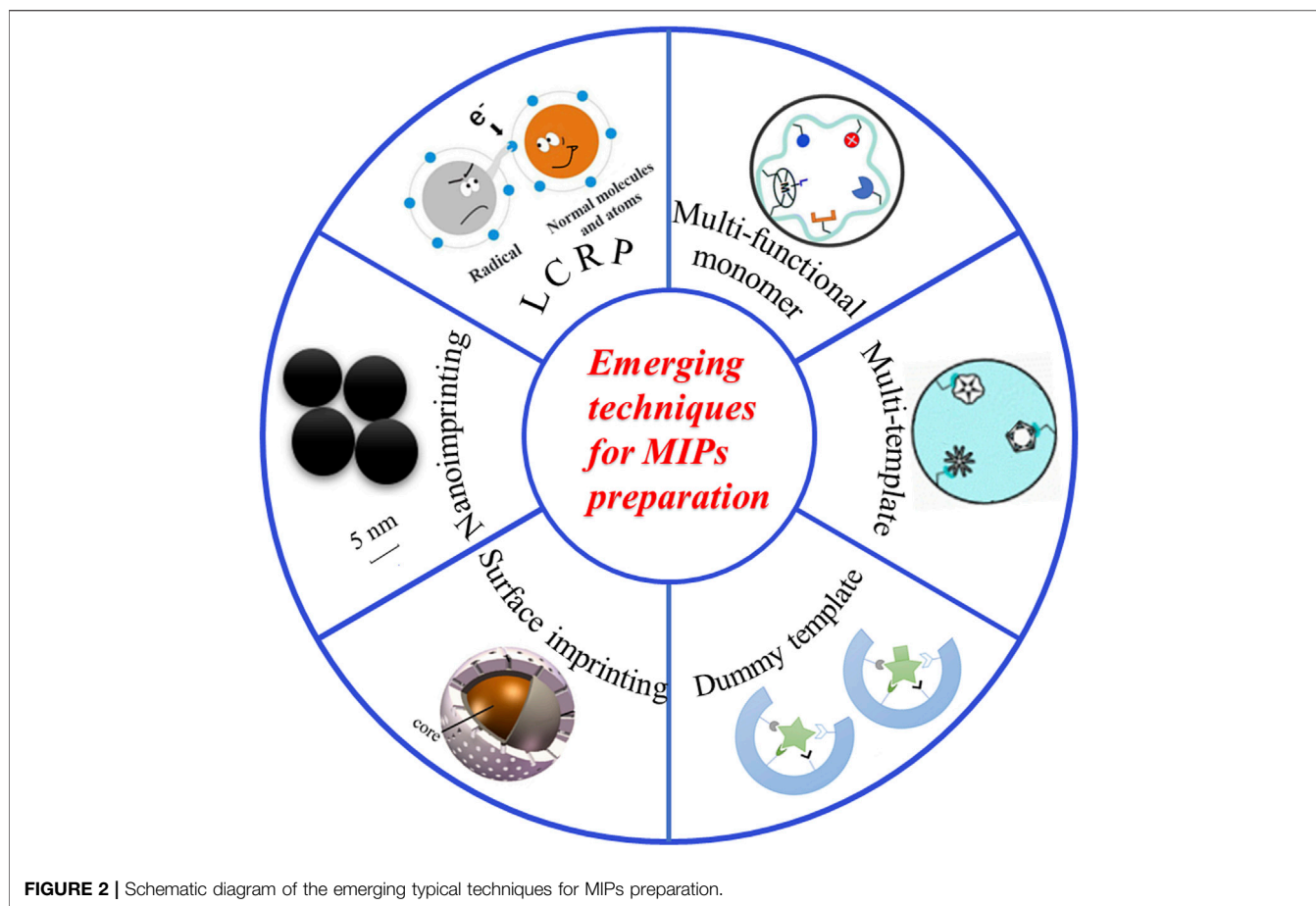


FIGURE 2 | Schematic diagram of the emerging typical techniques for MIPs preparation.

for detecting antibiotic residues (Ji et al., 2018; Liu et al., 2018; Negarian et al., 2019; Qin et al., 2019; Zhu et al., 2019a).

Nanoimprinting

Nanoimprinting technology is the technique of preparing nanosized MIPs. Nanomaterials have a large surface area and more binding sites, which can effectively improve the binding capacity of MIPs. To a certain extent, the problems of fewer target sites and low mass transfer rate of large-size MIPs are solved. Moreover, the nanostructured MIPs can be directly used without grinding, simplifying the experimental operation. The commonly used methods to synthesize nano-MIPs microspheres include precipitation polymerization (Liu et al., 2018), sol-gel (Li G. et al., 2018; Diaz-Alvarez et al., 2019), and core-shell polymerization (Qin et al., 2019). Nanoimprinting technology can be divided into zero-dimensional, one-dimensional, and two-dimensional types (Li and Wang, 2020).

LCRP

LCRP overcomes the disadvantage that the growth rate of the traditional radical polymerization chain is not easy to control and causes the particle size distribution of the polymer to be in a narrow range. Among them, atom transfer radical polymerization (ATRP) and reversible addition-fragmentation chain transfer (RAFT) polymerization are the most commonly used ones.

LCRP technology has been increasingly used for the determination of antibiotic residues (Liang Y. et al., 2018; Zhao et al., 2018; Cai et al., 2019; Li et al., 2020b; Cai et al., 2021) and other environmental contaminants (Azizi et al., 2020).

Multitemplate Imprinting

Multitemplate imprinting strategy means that two or more targeted analytes are as templates to prepare MIPs and thereby there are multiple recognition sites of template molecules in one imprinted polymer material (Wang et al., 2019). Due to the expansion of binding sites and recognition ability of MIPs, simultaneous recognition, enrichment, and separation of multiple targets can be realized, which greatly saves time and improves the utilization efficiency of MIPs. It has great potential in multiresidue and high-throughput analysis of complex samples (Wei et al., 2016; Xu et al., 2018; Lu et al., 2019; Dil et al., 2021).

Multifunctional Monomer Imprinting

The multifunctional monomer imprinting strategy is to take advantage of two or more functional monomers to interact with template molecules; by giving full play the synergistic effects of multiple functional monomers, MIPs selectivity is significantly enhanced and thereby enrichment ability. The strategy has been increasingly applied in antibiotics determination (Li G. et al., 2018; Li Z. et al., 2018; Cai et al., 2021).

Dummy Template Imprinting

Dummy template imprinting strategy is also called pseudotemplate imprinting strategy, which uses similar compounds to the target compounds as a template in the shape, size, structure, and functionality. It is a high requirement and especially suitable for the target compounds that are costly, flammable, explosive, easy to degrade, and having too low solubility. The strategy can effectively avoid template leakage pollution or inaccurate results. The most common method is the computer-aided design of dummy template toward MIPs (Song et al., 2017). MISPE prepared by dummy template imprinting strategy is widely used in the detection of antibiotic residues (Song et al., 2019; Zhang Z. et al., 2020; Gao et al., 2021).

APPLICATIONS OF MISPE TO ANALYSIS OF ANTIBIOTICS RESIDUES

The wide applications of MISPE to the analysis of antibiotics residues are summarized, including SAs, QNs, TCs, and other antibiotics. Some typical examples are listed in **Table 2**.

SAs

SAs residues are closely related to food and environmental safety levels. The pollution sources of SAs antibiotics mainly include medical sources (patients feces and urine, antibiotics remaining on medical supplies, losses in the production of antibiotics by pharmaceutical enterprises, etc.), animal sources (animal excrement and urine, leakage of sewage from farms, etc.), and aquaculture (overuse of antibiotics in the process of farming, etc.) (Zhang Y. et al., 2020). The key to detection is to develop fast, efficient, and highly selective pretreatment methods. Common SPE materials for SAs include HLB, C18, and Oasis MCX which are relatively general and commercially available, but they lack selectivity for SAs. MISPE can identify, extract, and enrich the target substances with high selectivity and specificity and has high adsorption capacity and stability. At the same time, various SPE devices have become smaller and are easier to operate. MISPE has been widely used coupled with chromatographic determination toward SAs in complex samples (Wang J. et al., 2018; Fonte et al., 2018; Huang et al., 2018; Kechagia et al., 2018; Xu et al., 2018; Zhao et al., 2018; Zhu et al., 2019a; Zhu et al., 2019c; Gao et al., 2021; Zhao et al., 2021).

In order to improve the selective adsorption performance of MIPs in strong polar solvents, Zhu et al. (2019a) synthesized sulfamethoxazole (SMZ) imprinted polymers in methanol by using a new ionic liquid (IL) functional monomer, namely, 1-butyl-3-vinylbromidazole (BVIM-Br). The MIPs exhibited highly specific recognition properties toward the three commonly found SAs (SMZ, sulfamonomethoxine (SMM), and sulfadiazine (SDZ)) in methanol, while low adsorption ability was displayed for the interferents. Then, a MISPE methodology was developed and successfully applied to effective enrichment of trace SMZ in soil and sediment samples, followed by HPLC analysis. The limits of detection (LODs) were all as low as 5 µg/L. The present research can offer

an important reference for influential MIPs preparation in aqueous media. Furthermore, Zhu et al. (2019c) prepared SMM surface imprinted polymers in the strong polar solvent of methanol with 1-allyl-3-vinylimidazolium chloride (AVIM-Cl) IL as a functional monomer. The developed MISPE coupled with HPLC was established for the selective extraction and sensitive determination of trace SMM in soil and sediment samples. The recoveries were high to 95.0–105.0% and the LOD was low to 1.0 µg/L. Such MIPs materials have a broad application prospect in the pretreatment of various complex samples.

Zhao et al. (2021) synthesized hydrophilic magnetic MIPs (HMMIPs) on the surface of silanated Fe₃O₄ via surface imprinting technique and using SDZ as a template molecule and employed them as DSPE adsorbents for the enrichment and purification of 10 SAs prior to HPLC-UV detection in chicken, cow milk, and goat milk samples. Under the optimal experimental conditions, good linearity in the range of 5 µg/L to 10 mg/L was exhibited, low LODs ranged from 0.57 to 1.50 µg/L, and high spiked recoveries were between 85.09 and 110.93%. The HMMIPs-DSPE method can provide a potentially applicable way for the sensitive, reliable, simple, and rapid detection of various drug residues. In the previous work (Zhao et al., 2018), water-compatible MIPs were also prepared by combining RAFT with reflux precipitation polymerization (RPP), and the resulting MISPE coupled with HPLC-MS/MS succeeded in the enrichment and determination of six SAs in animal-derived foods and water samples.

Gao et al. (2021) fabricated magnetic carbon nanotube dummy MIP (MCNTMIP) nanocomposite by surface imprinting technique and used it as MSPE adsorbent to realize the simultaneous separation and enrichment of 13 SAs (SDZ, sulfathiazole (ST), sulfamerazine (SM1), sulfamethazine (SM2), sulfamethizole (SMZO), SMM, sulfachloropyridazine (SCP), sulfadoxine (SO), SMZ, sulfisoxazole (SFZ), and sulfaquinoxaline (SOX), sulfadimethoxine (SDM), and avermectin B1a) in fish and shrimp samples. **Figure 3** illustrates the process of MIPs preparation and MSPE application. The MCNTMIP displayed a simple magnetic separation, specific molecular recognition, and high adsorption capacity. Coupled with ultra-high-performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS) determination of all the SAs, the LODs were all as low as 0.1 µg/kg, and the recoveries were in a range of 90.2–99.9%. Moreover, the precision values ranged from 0.5 to 9.1%. Consequently, the developed MCNTMIP-MSPE method can be routinely utilized for the trace analysis of SAs to ensure food quality and consumer safety.

QNs

QNs are a class of antibacterial drugs with 1,4-dihydro-4-oxyquinoline-3-carboxylic acid structure. They are widely used in clinical diagnosis and treatment, animal disease prevention, and growth promotion because of their advantages such as wide antibacterial spectrum, good efficacy, small toxic and side effects, simple synthesis, and low cost. The residual concentration of QNs is low, mostly ng/L or ng/kg. Excessive or improper use of QNs in animal-derived food has potential carcinogenic, teratogenic, and

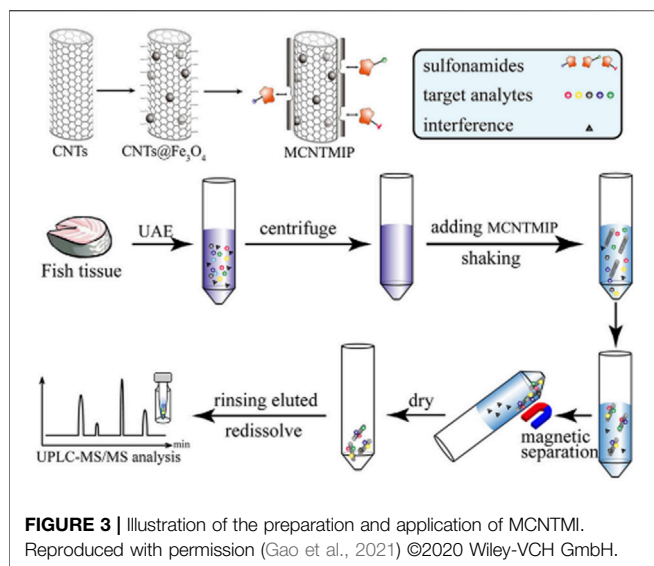
TABLE 2 | Applications of MIPs for SPE of antibiotics coupled with chromatographic determination.

SPE mode	Analyte	Template	MIPs preparation technique	Functional monomer	Cross-linker	Real sample	Detection technique	LOD	Ref.
(Conventional) SPE	SAR	SAR	ATRP	4-VP-co-MAA	EGDMA	Egg	UPLC/MS-MS	1.27 ng/g	Cai et al. (2021)
	SMZ	SMZ	Dual-functional monomer Surface imprinting	BVIM-Br	EDMA	Cultivated soil, vegetable soil, and sediment	HPLC	0.005 mg/L	Zhu et al. (2019b)
	OFX, GAT, NOR, CIP, DIF, PEF, SAR, ENX, floxacin, ENR, and LOM	–	–	–	–	Wastewater and sludge	UPLC/MS-MS	6–150 ng/L	Yu et al., 2020 ^a
	NFX	NFX	Noncovalent surface imprinting	MAA	EGDMA	Seawater	HPLC-DAD	2 µg/L	Qin et al. (2020)
DSPE	TC, CTC, and DC	TC, CTC, and DC	<i>In situ</i> polymerization	MMA	EDMA	Marine and sediments	HPLC-DAD	5 µg/kg	Huang L. et al. (2019)
	CIP and LEV	CIP and LEV	Bulk polymerization	[C2min] [Br]	EDMA	Egg	HPLC-FLD	3.0–5.5 µg/kg	Ma and Row (2019)
	NOR and ENR	NOR and ENR	Dual-template imprinting	MAA	EGDMA	Human urine	HPLC	0.06 and 0.27 µg/ml	Lu et al. (2019)
	NOR and ENR	NOR and ENR	Precipitation polymerization	MAA	EGDMA	Lake water	HPLC	0.22 and 0.36 µg/L	Lu et al. (2019)
DSPE	AZI, TYL, SPM, TIL, ERY, CLA, and ROX	TUL	Dual-template imprinting	MAA	EGDMA	Tap water	LC-MS/MS	0.2–0.5 µg/kg	Song et al. (2018)
	SDZ, STZ, SMR, SM2, SMT, SMP, SCP, SMM, SIA, and SDX	SM2	Precipitation polymerization	MAA	HEMA	Pork	HPLC	0.57–1.50 µg/L	Zhao et al. (2021)
	AZI, SPM, TIL, TYL, CLA, ROX, and JOS	SPM	Surface imprinting	MAA	EDMA	Chicken	HPLC-MS/MS	3–17 ng/kg	Ji et al. (2018)
	TC, OT, CTC, and DC	TC	Precipitation polymerization ^(a)	MAA	EGDMA	Cow milk	LVSS-CE	–	Aguilar et al., 2020
MSPE	OFX, PEF, NOR, ENR, and GAT	OFL	RAFT	MAA	EGDMA	Goat milk	HPLC-UV	1.02–3.15	Li et al. (2020b)
	SDZ, STZ, SMR, SM2, SMM, SCP, SO, SMZ, SFZ, SOX, SDM, and avermectin B1a	Sulfabenzamide	Surface imprinting	MAA	EGDMA	Milk	HPLC-UV	0.93–2.87 µg/L	Gao et al. (2021)
	CAP	CAP	Dummy template	MAA	EGDMA	River water	UPLC-MS/MS	0.1 µg/kg	Gao et al. (2021)
	CAP	CAP	Microwave-heating induced polymerization ^(b)	4-VP	TRIM	Fish and shrimp	UPLC-MS/MS	0.12 mg/kg	Kunsa-Ngiem et al. (2018)
TC and OT	CAP	CAP	Surface imprinting	MAA	EGDMA	Chicken feed	UPLC-MS/MS	0.12 mg/kg	Kunsa-Ngiem et al. (2018)
	TC	CAP	Suspension polymerization	MAA and/or AM	EGDMA	Pork and honey	HPLC-UV	10 µg/L	Li Z. et al. (2018)
	TC	Pickering emulsion polymerization	Surface imprinting	MAA	EGDMA	Marine sediments	HPLC-DAD	0.1 µg/L	Qin et al. (2019)
	TC	Surface imprinting	MAA	DVB	Fish, chicken, and tap water	HPLC	HPLC	1.42 and 1.58 µg/L	Ma et al. (2020)
PZQ enantiomers	PZQ	Surface imprinting	Py	–	Sheep milk	HPLC-DAD	0.01 µg/ml	Nascimento et al. (2020)	
TC, OT, and CTC	TC	Nanoimprinting	Surface imprinting ^(c)	APBA	APBA	Milk powder	UPLC-MS/MS	0.278, 0.318, and 0.217 ng/g	Wang S. et al. (2018)
SPME	DAN, NOR, ENR, and CIP	ENR	–	MAA	EGDMA	Surface water, groundwater, and urine	HPLC-MS/MS	0.1–10 µg/L	Barahona et al. (2019)
	ERY, CLA, AZI, LEU, and ROX	ROX	Surface imprinting	MAA	EGDMA	Drinking water, honey, and milk	ESI-MS	0.003–0.05 1.1–5.1 1.9–15.8 ng/g	Liu et al. (2019)
SBSE	SDZ, SMD, SMM, and SDM	SMM	Emulsion polymerization	MAA	EGDMA	Regular chicken feed, and pig feed	HPLC-MS/MS	1.5–3.4 µg/L	Cui et al. (2021)
PT-SPE	CIP and ENR	ENR	Thermal radical polymerization ^(d)	MAA	EGDMA	Suckling pig feed	HPLC-MS/MS	1.5–3.4 µg/L	Cui et al. (2021)
	MARBO and NOR	ABA	–	1-VI	TRIM	Two types of laying-hen feeds	HPLC-DAD	18 ng/ml	de Oliveira et al. (2016)
	ABA and EPR	ABA	–	1-VI	TRIM	Human urine	HPLC-DAD	18 ng/ml	de Oliveira et al. (2016)
	MOX	CIP	<i>In situ</i> polymerization	MAA	EGDMA	Mineral water grape juice	HPLC-UV	–	Teixeira et al. (2018)
	CIP	CIP	<i>In situ</i> polymerization	MAA	EGDMA	Seawater and human blood plasma tablet	Spectrophotometry	1.50 µg/L	Hashemi et al., 2019

AM, acrylamide; APBA, 3-aminophenylboronic acid; AZI, azithromycin; CLA, clarithromycin; [C2min][Br]: 1-vinyl-3-ethylimidazolium bromide; DAN, danofloxacin; DIF, difloxacin; DVB, divinylbenzene; EDMA, ethylene dimethacrylate; EGDMA, ethylene glycol dimethacrylate; ERY, erythromycin; LEU, leucomycin; LEV, levofloxacin; HEMA, hydroxyethyl methacrylate; IVM, ivermectin; Py, pyrrole; ROX, roxithromycin; SDX, sulfameter; SIA, sulfisoxazole; SMP, sulfamethoxy-pyridazine; SMD, sulfameter; SMT, sulfamethoxydiazine; MAA, methacrylic acid; MMA, methyl methacrylate; SPM, spiramycin; TIL, tilimicosin; TYL, tylosin; 4-VP, 4-vinyl pyridine.

(a) Persulfate; (b) 1,1-azobis(cyclohexanecarbonitrile) (ABCN); (c) 0.1 mol/L Na₂S₂O₈ (6.5 ml) aqueous solution; (d) 4,4'-azobis(4-cyanovaleric acid), four different initiators; the other initiators were AIBN.

^aMIP-SPE cartridges (SupelMIP) were obtained from Sigma-Aldrich (Shanghai, China).



mutagenic effects, thus threatening human health. Almost all countries have formulated the maximum residual limit standard (MRLS) for quinolone antibiotics. MISPE is widely used in the determination of QNs in complex substrates (Li G. et al., 2018; Wang J. et al., 2018; Hu et al., 2018; Rodriguez-Gomez et al., 2018; Barahona et al., 2019; Zhu et al., 2019b; Ma and Row, 2019; Li et al., 2020b; Tian et al., 2020; Yu et al., 2020; Cai et al., 2021; Soares et al., 2021).

Our group (Lu et al., 2019) prepared novel double-template MIPs (dt-MIPs) via a simple and facile precipitation polymerization method with norfloxacin (NOR) and enrofloxacin (ENR) as templates and used them in DSPE combined with HPLC-DAD for the simultaneous selective enrichment and determination of two fluoroquinolones (FQs) in environmental water samples. **Figure 4** schematically shows the dt-MIPs preparation and DSPE process. The well-prepared dt-MIPs exhibited good adsorption capacity and selectivity for NOR and ENR, with high enrichment factors of 71 and 61, respectively. Good linearity was obtained in the range of 1–200 µg/L. The LOD and limit of quantification (LOQ) values for NOR were 0.22 and 0.67 µg/L, respectively, and 0.36 and 0.98 µg/L for ENR. Satisfactory recoveries of the two FQs were attained of 80.9–101.0% with relative standard deviations (RSDs) of 0.9–6.9% from spiked lake, sea, and tap water samples. This study not only offered a good method choice for FQs detection but also enriched the research connotation of multitemplate imprinting.

Yu et al. (2020) developed a MISPE method for the simultaneous enrichment of 11 FQs [ofloxacin (OFX), gatifloxacin (GAT), NOR, ciprofloxacin (CIP), difloxacin, pefloxacin (PEF), sarafloxacin (SAR), enoxacin (ENX), floxacin, ENR, and lomefloxacin (LOM)] in water by UPLC/MS-MS determination. The attained LODs of FQs were within 6–150 ng/L. The recoveries of all the targeted FQs in sample matrices were higher than 75%, and RSDs were below 15%. The developed MISPE-UPLC/MS-MS proved to be effective for the determination of FQs in wastewater and sludge samples.

Li et al. (2020b) successfully prepared restricted-access media-imprinted nanomaterials (RAM-MIPs) on the surface of the

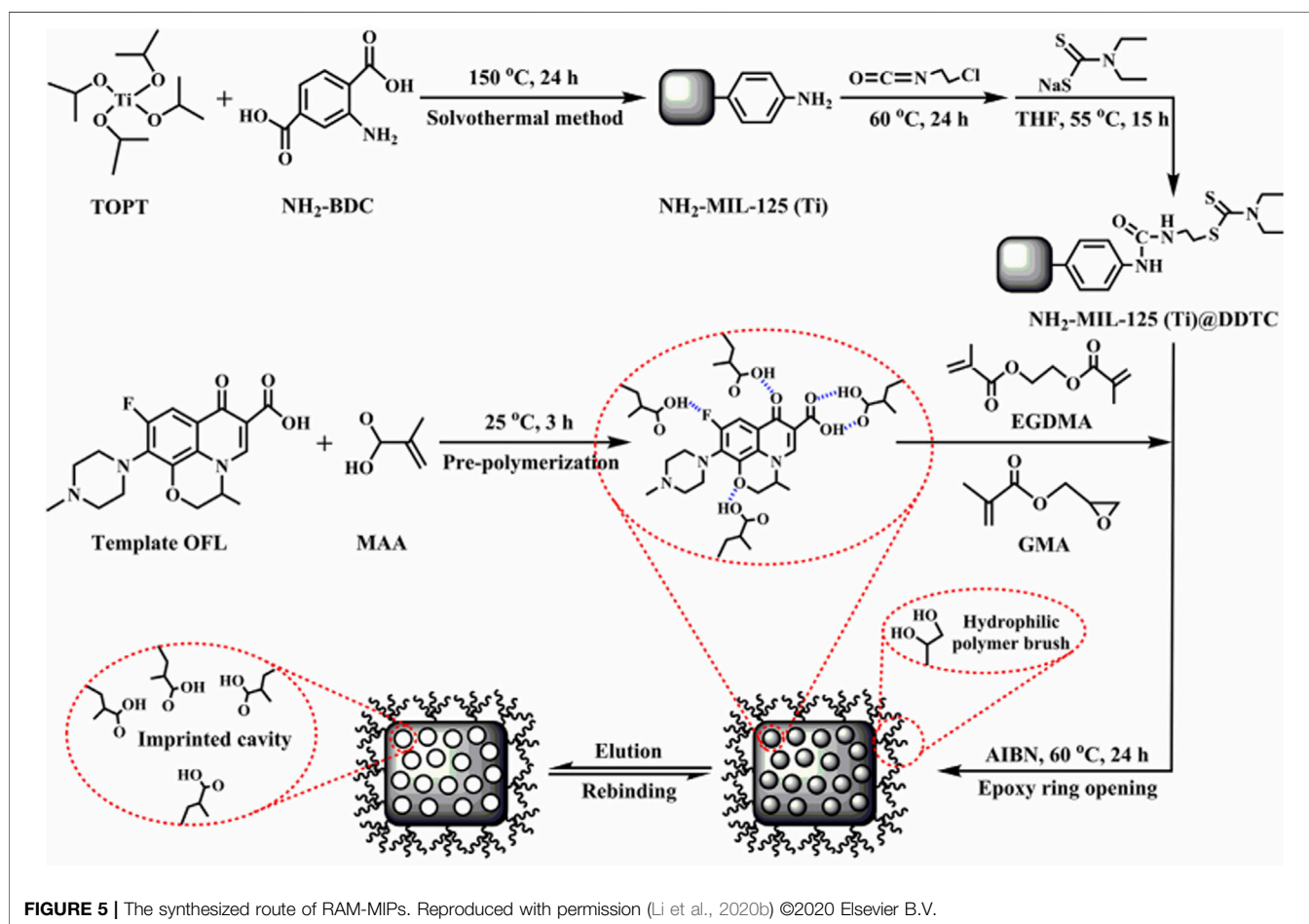
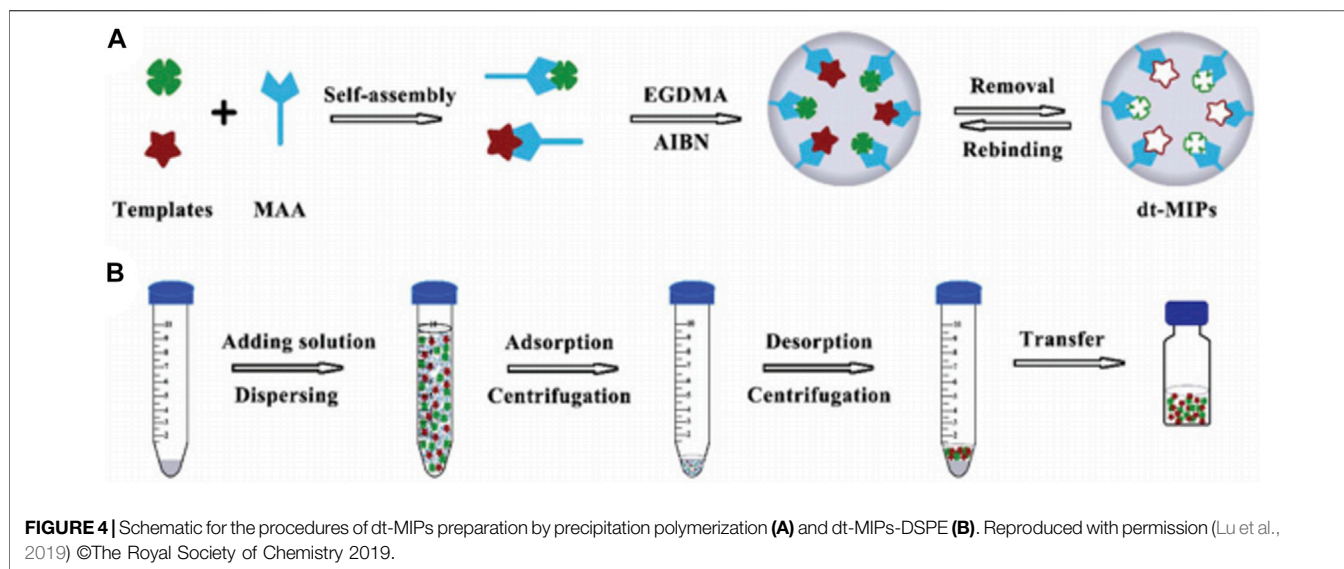
metal-organic framework (MOF) by RAFT. **Figure 5** shows the synthesized route of the RAM-MIPs. Then, they were applied for the DSPE of five FQs (OFX, PEF, NOR, ENR, and GAT) in milk and river water samples prior to HPLC-UV detection. The method attained low LODs, namely, 1.02–3.15 µg/L for milk samples and 0.93–2.87 µg/L for river water samples, respectively, as well as satisfactory recoveries, namely, 80.7–103.5% and 85.1–105.9%, respectively. In comparison with other materials, the RAM-MIP materials are significantly advantageous owing to their simple preparation conditions, uniform and controllable imprinting layer thickness, fast adsorption rate, and so on. The present study demonstrates that RAM-MIP (prepared with MOF as a matrix)-based SPE has broad prospects toward efficient extraction of trace FQs in complex samples.

Cai et al. (2021), using surface-initiated ATRP and sarafloxacin (SAR) as a template, constructed monodisperse RAM-MIPs, and three methods were adopted, as illustrated in **Figure 6**. The optimum synthesis method was to combine 4-vinylpyridine (VP) and methacrylic acid (MAA) (1:3) as monomers and to select an 8:1:32:8 ratio of a template molecule, cross-linker, and restricted-access functional monomer. The RAM-MIPs showed a high IF (6.05) and the selectivity coefficients were 1.86–2.64 between SAR and other FQs. The RAM-MIP-packed SPE showed higher enrichment ability toward SAR in a complex protein-containing solution than that of traditional MIP-packed one. As a result, the MISPE coupled with the HPLC-UV method achieved a low LOQ for SAR at 4.23 ng/g and the high mean recoveries within 94.0–101.3%. The present study indicated a great application potential of the RAM-MIPs based SPE for trace analysis in complex samples. Furthermore, the proposed functional monomer ratio and rebinding method opened a new way for devising and synthesizing various MIPs.

de Oliveira et al. (2016) used ENRO as the template molecule to synthesize MIP1, adopted a multitemplate imprinting strategy (four studied FQs as the template molecules) to synthesize MIP2, and utilized it for the simultaneous PT-SPE of the four FQs (CIP, ENR, marbofloxacin (MARBO), and NOR) in human urine samples. By comparison, MIP1 proved a better adsorbent, and high extraction efficiency was obtained to ENRO (96.0%). **Figure 7** schematically shows the apparatus employed for PT-MIP-µ-SPE. It was possible to extract CIPRO (~40%), NOR (~40%), and MARBO (~80%) due to the similarity of the molecular structures. The method attained good linearity from 39 to 1,260 ng/ml for individual FQ, and the LOQ for individual FQ was as low as 39 ng/ml. Finally, the validated PT-MIP-µ-SPE method was proved to be practically applicable, through the preliminary cumulative urinary excretion study after administrating CIPRO to a healthy volunteer.

TCs

TCs, as a kind of broad-spectrum antibiotics produced by *Streptomyces*, have caused serious harm to the ecological environment and human health because of their wide use and hence residues. Huang L. et al. (2019) used TC, chlortetracycline (CTC), and doxycycline (DC) as the templates and magnetic graphene oxide (Fe₃O₄/GO) as the supporting material to prepare



magnetic multi-MIPs. Then chip-based magnetic multi-MIPs monolithic capillary array columns were constructed for simultaneous MSPE and determination of the TCs in eggs

samples. High affinity and specificity to TC, CTC, and DC were shown and the IFs reached 86–104-fold. The LOD values ranged from 3.0 to 5.5 µg/kg. Therefore, the MISPE columns

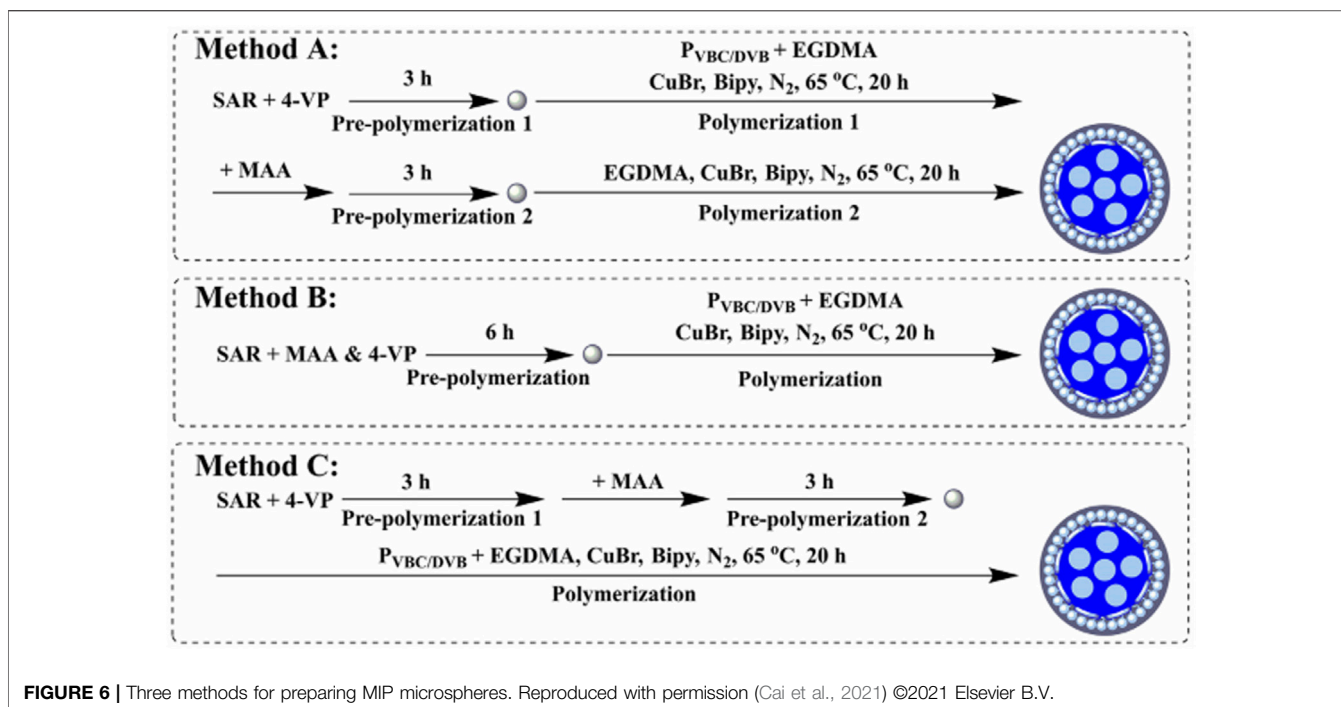
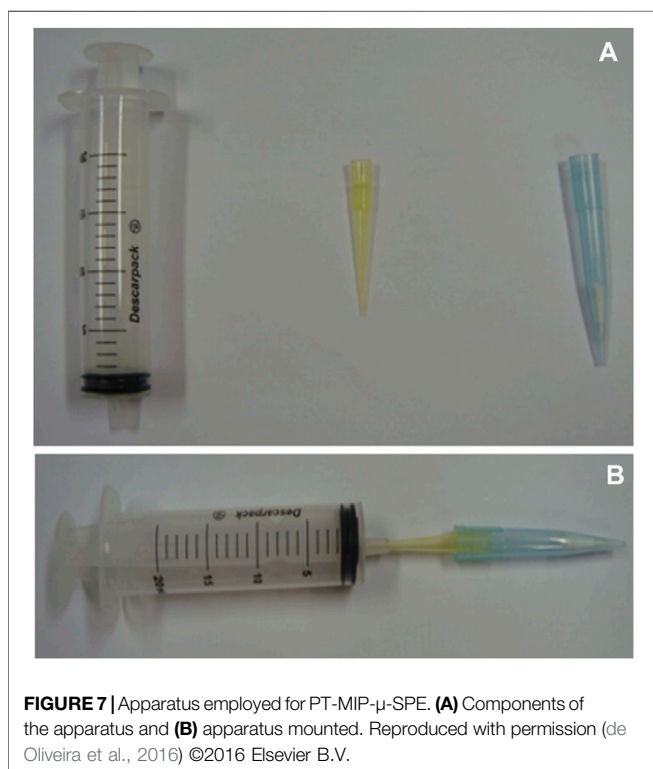


FIGURE 6 | Three methods for preparing MIP microspheres. Reproduced with permission (Cai et al., 2021) ©2021 Elsevier B.V.



afforded hopeful perspectives for the facile extraction of antibiotics from complicated samples.

Aguilar et al. (2020) synthesized the MIPs by precipitation polymerization using TC as template molecule and applied them

for dispersive SPME (DSPME) and removal of TC residues in milk samples. The molecular recognition properties and selectivity of MIPs against four TCs (TC, oxytetracycline (OT), CTC and DC) were evaluated, and then high selectivity was demonstrated for the four TCs. The MIP-based DSPME process provided a high removal ratio between 81.83 and 96.44% with RSD < 5% in all cases. Compared to classical removal methods, the present method was faster and required lower solvent consumption and minimum sample manipulation. Therefore, a promising prospect can be expected for facily synthesizing efficient and selective adsorbents and utilizing MISPE for the simultaneous removal of multiple contaminants residues.

Ma et al. (2020) prepared magnetic molecularly imprinted biochar microspheres with specific adsorption of TCs (TC; OT) by Pickering emulsion polymerization. The obtained materials were employed as adsorbents for extraction and purification of TCs in actual samples (fish, chicken, and tap water). This method was simple in preparation process and cost-effective; the synthesized polymer was a regular spherical structure with magnetic response characteristics, which can simplify the extraction and purification of sample pretreatment. It offers a new idea for the application of MIPs based biochar in contaminant detection in food samples.

Other Antibiotics

CAP is a kind of broad-spectrum antibiotics isolated from *Streptomyces venezuelae*. Because its long-term and high dose use easily caused granulocytosis, aplastic anemia, and other diseases, China has banned its use in feed-animals (especially laying hens and dairy cows) and required CAPs residues to be mandatory test items in all aquatic products, livestock, and poultry products (Zhang et al., 2021). BLAs are a broad class of antibiotics,

and their residues mainly come from agricultural and veterinary drugs, medical drugs, and wastewater treatment discharges from sewage plants. They pose potential threats to the human body and ecology. For example, a few patients will have allergic reactions to BLAs, and the residual antibiotics in the soil will affect the growth of plant roots and seed germination, etc. In addition, the pollution of Antibiotic-Resistant Bacteria (ARB) and Antibiotics Resistance Gene (ARG) caused by the abuse of BLAs and other antibiotics is threatening human health and ecological safety (Li et al., 2020a). MALs are widely used in clinical and veterinary medicine fields, with a broad spectrum of antimicrobial effects, especially for animal husbandry and aquaculture (Liang and Zhang, 2021). At present, more and more efforts have been devoted to the monitoring of MALs residues.

Using MISPE technology with excellent performance, the trace detection of various antibiotics in diverse complex substrates has been realized (Li Z. et al., 2018; Lian and Wang, 2018; Xie et al., 2018; Negarian et al., 2019; Qin et al., 2019; Garza Montelongo et al., 2020; Tan et al., 2020). For example, Qin et al. (2019) developed a straightforward method for selective separation of CAP from marine sediment samples. CAP-MMIPs were synthesized via surface imprinting and nanoimprinting technologies. The material has a perfect core-shell structure, excellent thermal stability, and high selectivity toward CAP. The CAP-MMIPs were employed for fast and selective SPE of CAP followed by HPLC-DAD. An excellent linearity was attained from 0.1 to 20 mg/L ($R^2 = 0.999$, $n = 3$), and the LOD was 0.1 $\mu\text{g/L}$. The spiked recoveries were between 77.9 and 102.5% with low RSDs ($< 6.3\%$). Good reusability was achieved (at least 5 times) by the regeneration and there was hardly any loss of selectivity and adsorption capability. Such MMIPs-SPE method can provide a vital alternate to traditional extraction ones for preparing environmental samples.

Teixeira et al. (2018) prepared 1-vinylimidazole-co-trimethylolpropane trimethacrylate- (1-VI-co-TRIM-) based MIPs adsorbent for PT-SPE of abamectin (ABA), eprinomectin (EPR), and moxidectin (MOX), coupled to HPLC-UV determination. The performance criteria for linearity, sensitivity, precision, accuracy, recovery, robustness, and stability were systematically assessed and were within the recommended guidelines. The validated PT-MIP-SPE proved to be economical, simple, easy-to-perform, and potentially applicable for the extraction of MALs in complicated samples.

Nascimento et al. (2020) firstly synthesized a new selective adsorbent based on magnetic molecularly imprinted polypyrrole-conducting polymer (MMIPPy) and applied it to the MSPE of praziquantel (PZQ) enantiomers [(R)-(-)-PZQ and (S)-(+)-PZQ] combined with HPLC-DAD determination. Under optimal conditions, excellent linearity was attained in a range of 0.01–10 $\mu\text{g/ml}$, with correlation coefficients higher than 0.98 and RSDs less than 15%. The LOQ was 0.01 $\mu\text{g/ml}$ for both enantiomers, and RSDs and relative errors were below 20%. The method was applied satisfactorily for the determination of PZQ enantiomers from sheep milk samples with the possibility to other analytes in different complex matrices. The economical, simple, and easy-to-perform MMIPPy-MSPE method suggested great application potential for antibiotics residues determination.

CONCLUSIONS AND PROSPECTS

To summarize, we review recent advances on both the classification of MISPE and new imprinting techniques for antibiotics coupled with chromatographic analysis, with emphasis on typical examples of MISPE in SAs, QNs, TCs, and other antibiotics. The use of emerging typical imprinting techniques has greatly improved the performance of MISPE and further broadened its application scope. The advancement of new imprinting techniques, especially the combination of multiple techniques, can effectively solve some problems in the practical application of traditional MIPs, such as low binding capacity, template leakage, and difficulties in aqueous phase identification, and provide a better performance, easier separation, and intelligently controlled release of MIPs as solid-phase sorbents. This can provide an effective means to eliminate matrix interference and enrich trace antibiotics with high selectivity. Moreover, the sample pretreatment will definitely develop in the direction of economical, efficient, environmentally friendly, and easy to operate related aspects. In order to further improve the extraction efficiency of MISPE by using ideal MIPs with greater adsorption capacity, higher selectivity, better hydrophilicity, and easier separation, it is necessary to introduce more and advanced preparation techniques, especially to explore the rational synergistic combination of multiple imprinting techniques. Furthermore, the large-scale production and commercialization of the well-prepared MIPs and well-established MISPE should be given more attention, in order to push forward their greater advance and wider applications.

AUTHOR CONTRIBUTIONS

DS: writing the original draft. ZS: supervision, revising the manuscript, and funding support. YZ: searching for references and writing part of the original draft. YW: reviewing the manuscript. ML: reviewing and revising the manuscript and funding support. HL: reviewing and revising the manuscript. LW: searching for reference and revising the manuscript. JL: supervision, revising the manuscript, and funding support. LC: editing the manuscript and funding support.

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