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Synthesis and characterization of a molecularly imprinted polymer for the determination of trace tributyltin in seawater and seafood by liquid chromatography–tandem mass spectroscopy

Shanshan Zhu[a], Futao Hu[a], Ting Yang[c], Ning Gan[b,*], Daodong Pan[a,**], Yuting Cao[b], Dazhen Wu[b]

[a] Faculty of Marine Science, Ningbo University, China
[b] Faculty of Material Science and Chemical Engineering, Ningbo University, China
[c] Ningbo Agricultural Science Research Institute, Ningbo, China

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ABSTRACT

Analysis of tributyltin chloride (TBT) in environmental samples, such as seawater, is important in order to evaluate the TBT contamination and accumulation in the trophic chain. The environmental impact of organotin compounds has been a particular focus of analytical studies. The present study reports the use of molecular imprinting technology coupled with liquid chromatography–tandem mass spectrometry (LC–MS/MS) to determine trace amounts of TBT in seawater and seafood (mussel tissue samples). The imprinted polymer was synthesized by a non-covalent free-radical approach using acrylamide (AM) as a monomer and TBT as a template molecule in acetonitrile solvent (polymerization media). The imprinted polymer synthesized by this approach exhibited good adsorptive capacity and allowed specific retention of TBT. Recoveries of TBT in seawater samples spiked with different TBT concentrations ranged from 67.2% to 81.1% with peak area precision (RSD) < 3.7%, and recoveries of TBT in mussel tissue samples ranged from 75.0% to 94.2% with RSD < 4.8%.

1. Introduction

Extensive use of organotins as biocides in antifouling paints and as fungicides in agricultural activities results in the direct release of these compounds into the aquatic environment. High concentrations of organotins have been detected in water, sediments, and seafoods, thus posing an ecotoxicological threat to non-target organisms [1]. Even very low concentrations of organotin compounds in the environment may result in harmful effects on marine organisms. Different compounds have different environmental effects and toxicities [2]. Organotins are synthetic organometallic compounds including monobutyltin dichloride (MBT), dibutyltin dichloride (DBT), tributyltin chloride (TBT) and triphenyltin chloride (TPhT) (Fig. 1). Among organotins, particularly TBT has been reported to have toxic effects on aquatic organisms [3], and is also one of the most toxic chemicals that have been released into the marine environment through an anthropogenic source [4]. It can cause high larval mortality, shell deformation, and retardation of cell growth [5,6]. Moreover, numerous deleterious biological effects of TBT on non-target organisms have been reported [7].

Organotin compounds (OTCs) have been found in the marine environment for a long time. Although their use in recreational watercrafts is now regulated in most countries, TBT is still employed in larger commercial vessels and is released in the natural waters during vessel maintenance activities [8]. As a result, TBT contamination has been reported in seawater, sediments, and aquatic organisms. Therefore, monitoring the levels of TBT and other OTCs in the marine environment is of primary importance.

Several analytical methods based on various separation and detection techniques have been developed to determine the presence of OTCs in the environment. The main separation techniques include gas chromatography (GC) and high-performance liquid chromatography (HPLC). And the main detection techniques include fluorescence detection, atomic emission spectrometry (AES), and mass spectrometry (MS) etc. [9,10]. However, the poor volatility of OTCs calls for a derivatization step prior to GC. In contrast, HPLC does not involve such a step, thus eliminating a potential source of analytical uncertainty in the final result [2]. Owing to this advantage, OTCs are quantified by means of HPLC coupled with various detectors. Among the detection techniques, tandem mass spectrometry (MS/MS) offers a number of advantages, such as more selective separation, element specificity, low detection limits, and high sensitivity [11], thus making it the most widely used technique for the quantitation of OTCs in the environment and food [12].

* Corresponding author. Tel.: +86 574 87608347; fax: +86 574 87608347.
** Corresponding author. Tel.: +86 574 87609987; fax: +86 574 87609987.
E-mail addresses: ganning@nbu.edu.cn (N. Gan), daodongpan@163.com (D. Pan).

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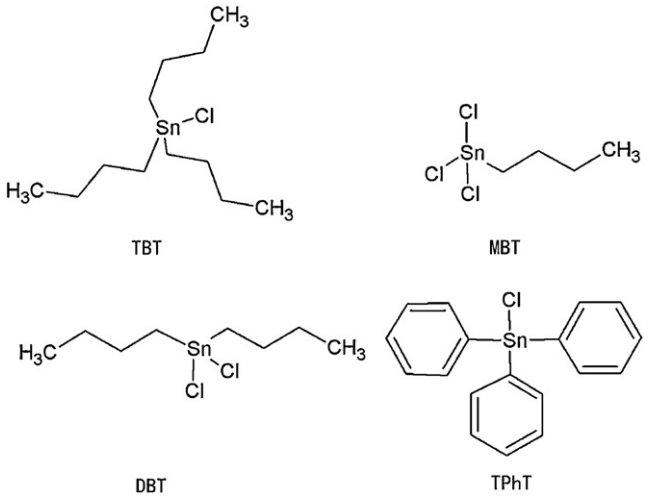

Fig. 1. Chemical structures of TBT and structurally related compounds MBT, DBT and TPhT.

Preconcentration techniques play an important role in the analysis of OTCs in samples. A sample preparation step prior as liquid–liquid extraction (LLE), solid-phase extraction (SPE), supercritical fluid extraction, microwave-assisted extraction and pressurized liquid extraction have been applied or recommended for the determination of OTCs [13]. However, they also have some shortcomings, such as time- consuming [14], labor-intensive, and hazardous to health or require highly qualified personnel and sophisticated instrumentation. For analysis of complex samples, such as biological samples, molecular imprinting technology can be used for sample preparation procedures with higher selectivity to enrich the target molecules from complex matrices. Molecular imprinting of synthetic polymers is a process in which a functional monomer and a cross-linking agent are copolymerized in the presence of a target analyte (the imprint molecule) that acts as a molecular template to imitate natural molecular recognition [15,16]. After the template is removed from the resulting polymer matrices, binding sites whose sizes and shapes are complementary to the template are generated [17]. Consequently, molecular imprinting has proven to be an efficient method to produce functionalized materials that have the ability to recognize a specific template from a mixture of closely related compounds [18]. In other words, the hypothetical "lock and key" relationship between an enzyme and its substrate can be applied to pursue similar synthetic approaches, with the aim of obtaining tailored binding materials by chemical means [19]. Therefore, molecularly imprinted polymers (MIPs) have applications in many areas, such as chiral separation, chemical sensors, and biosensors.

In the present study, an MIP was used as a sorbent and coupled with LC–MS/MS for the detection of trace amounts of TBT in seawater and seafood. We synthesized the MIP using the non-covalent molecular imprinting technique, with TBT as a template, acrylamide (AM) as a functional monomer, and ethylene glycol dimethacrylate (EGDMA) as a cross-linker agent in acetonitrile solvent. The synthesized MIP showed high selectivity, fast binding kinetics, and good adsorption properties.

2. Experimental

2.1. Instrumentation and reagents

The morphology of the polymer particles was examined using a transmission electron microscope (H-7650 Hitachi, Japan). The following instruments were used: the Anke TDL-5 centrifuge (Shanghai Anting Science Equipment Factory, China), which was used for the separation of preconcentration; a muller (ZHM1A, Beijing Zhonghe Venture Science Technology Development Factory, China), which was used for grinding of the polymer; an LC–MS/MS system, which consisted of an Acquity UPLC Xevo^{TM} TQ MS equipped with an electrospray ionization (ESI) interface (Waters, USA).

The compounds TBT (>96%), MBT (>96%), DBT (>97%), and TPhT (>95%) were obtained from the Jingchun Reagent Factory (Shanghai, China). Stock solutions of the standards (100 g L^{-1}) were prepared in acetonitrile and stored at −4 °C under dark conditions. The polymerization reagents AM, EGDMA, and azobisisobutyronitrile (AIBN) were all obtained from the Aladdin Chemistry Co., Ltd. (Shanghai, China). All reagents were of the highest available purity and at least analytical grade. All solutions used for LC–MS/MS were filtered through a nylon 0.45 μm filter prior to use. Fibers were provided by the Jiuwu Hitech Co., Ltd. (Shanghai, China).

2.2. Spiking of the seawater samples

Each seawater sample (50 mL) was transferred into a 100 mL conical flask, spiked with 100 μL of standard solution of TBT (50, 100, and 500 μg L^{-1}) containing 5.0, 10.0, and 50.0 ng of TBT, respectively. The spiked seawater samples were left overnight before the extraction with the polymers.

2.3. Spiking of the mussel tissue

Each frozen mussel tissue sample (0.1 g) was taken in a 10 mL centrifuge tube. It was spiked with 100 μL of standard TBT solution (50, 100, and 500 μg L^{-1}) containing 5.0, 10.0, and 50.0 ng of TBT, respectively, then shaken for 1 h and left overnight before performing the extraction procedure.

2.4. Extraction of TBT from the mussel tissue

Ultrasonication of the tissue sample was performed for 30 s at about 30 kHz using the 5 mL of extraction media [MeOH/acetic acid (HAc) 1:1, v/v]; followed by centrifugation at 4500 rpm for 2 min. The supernatant after centrifugation was saved for analysis. These above procedures were repeated [20]. The sample was extracted twice with the same extract to improve the recovery rate of TBT. Both extracts (10 mL) were combined, and then re-dissolved in 50 mL of water. The extracts were now ready for preconcentration.

2.5. Preparation of the polymers

The dispersed polyacrylamide microbeans was synthesized to the Chinese patent of which application No. 200510015571 [21], the TBT (1 mmol) was mixed with 4 mmol of the monomer (AM) in a 50 mL calibrated flask. The mixture was left for 5 min at room temperature for equilibration. Then, 20 mmol of the cross-linking agent, EGDMA, 15 mg of AIBN (as an initiator), and 5 mL of acetonitrile (as a solvent) were added. The mixture was incubated in a water-bath at 65 °C for 20 h in order to complete the polymerization process. The polymer was then ground with muller for aggregating in the polymerization and sieved to a size of ~500 nm, so the particle size is relatively uniform. The polymer was then washed using the Soxhlet extraction method [13] for 60 h with 0.3 M HCl in methanol (MeOH) to remove the template. Finally, the polymer was air-dried and stored until use. The non-imprinted polymer (NIP) was also prepared following a similar scheme but in the absence of the template.

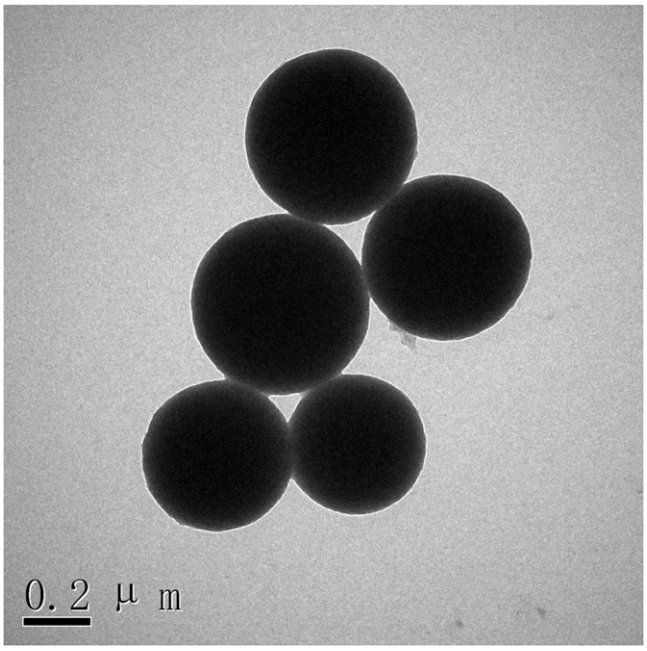

Fig. 2. Characterization of molecularly imprinted polymers (MIPs) by transmission electron microscopy (TEM).

2.6. Preconcentration and elution

Spiked seawater and spiked mussel tissue extracts containing TBT was passed over 20 mg of the polymers, and then stirred for 24 h at room temperature. After washing the polymers with 6 mL of 4:1 MeOH:CH_2Cl_2, the final elution of the retained compounds was carried out with 6 mL of 0.1 M formic acid in methanol under stirring for 2 h at room temperature. The supernatant was obtained by centrifugation at 5000 rpm for 5 min and dried under an N_2 stream. Finally, the residues were re-dissolved in 1 mL of mobile phase containing formic acid and water (1:9, v/v) for further LC–MS/MS analysis.

3. Results and discussion

3.1. LC–MS/MS parameter optimization

The LC–MS/MS analysis was performed using an Acquity UPLC-Xevo^{TM} TQ MS LC–MS/MS system equipped with an ESI interface (Waters, USA). The stationary phase was an Acquity BHE C_{18} 50 mm × 2.1 mm column (particle size, 1.7 μm) with a column temperature of set at 30 °C, and the mobile phase was formic acid and water (1:9, v/v). A flow rate of 0.2 mL min^{-1} was maintained for 10 min, following the injection of a 10 μL sample.

Ion source temperature was set at 150 °C, and capillary and the radio frequency (RF) lens voltages were set to 3.5 kV and 0.5 V, respectively. Nitrogen gas was used for shielding and drying, while argon gas was used as the collision gas.

Analyte determination was carried out in the multiple reaction monitoring (MRM) mode.

3.2. Transmission electron microscopy (TEM)

The micrograph in Fig. 2 shows the formation of spherical MIP particles. The diameters of the MIP particles were in the submicron range, and the particle size was about 500 nm.

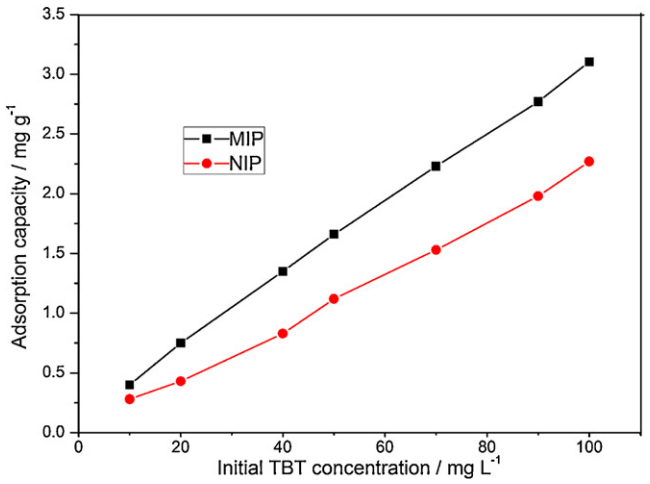

Fig. 3. Adsorption capacities of the MIPs and NIPs.

3.3. Adsorption capacity of the polymers

To measure the adsorption capacity of the polymers, 20 mg of the NIP or MIP particles were equilibrated with 10 mL of TBT, the concentration of which ranged from 10 mg/L to 100 mg/L dissolved in ethanol. The mixture was magnetically stirred for 24 h at room temperature and then separated by centrifugation at 5000 rpm for 5 min. The supernatant was analyzed for the un-extracted TBT by LC–MS/MS. The isothermal adsorptions of the polymers are plotted in Fig. 3.

It can be seen from Fig. 3 that the adsorption capacity of the MIPs and NIPs for the template molecule increases by increasing the initial concentration of TBT. Clearly, the adsorption capacity of the MIPs was higher than that of the NIPs which indicated that the MIPs exhibited a stronger memory function for TBT, and that the binding affinity of the MIPs was mainly from the specific sites formed by the imprinting effect. The RSDs were less than 4.3% and 7.6% at each concentration of MIPs and NIPs, respectively ($n = 3$).

The Langmuir and Freundlich models were used to describe the adsorption isotherms of TBT for the adsorbents (polymers) at 298 K. The Freundlich equation [22–24] is given as follows:

$$q_e = K_F C_e^{1/n} \quad \text{(nonlinear form)} \tag{1}$$

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad \text{(linear form)} \tag{2}$$

the Langmuir equation [22,25] may be written as

$$q_e = \frac{Q^0 b C_e}{1 + bC_e} \quad \text{(nonlinear form)} \tag{3}$$

$$\frac{C_e}{q_e} = \frac{1}{Q^0 b} + \frac{1}{Q^0} C_e \quad \text{(linear form)} \tag{4}$$

where q_e is the amount of TBT bound to the MIPs at equilibrium (mg/g), C_e is the free analytical concentration of solute at equilibrium (mg/L), K_F is a constant indicative of the relative adsorptive capacity of the adsorbent (mg/g), and the constant $1/n$ indicates the intensity of the adsorption, Q^0 is the apparent maximum adsorption capacity, and b is the dissociation constant.

As shown in Table 1, optimal fitting of the adsorption isotherm data to the Freundlich isotherm model is obtained. b and n represent isothermal parameters, which can be regarded as being representative of the strength of adsorption and the heterogeneity of the system [22], respectively. Therefore, the deviation of n from 1.0 may be taken as a measure of the deviation from the surface homogeneity. Hence, the fitting values of the Freundlich parameters and

Table 1
Fitting parameters for TBT adsorption on MIP.

Temperature	Freundlich model			Langmuir model		
	R^2	K_F	n	R^2	b	Q^0 (mg/g)
298 K	0.999	0.06	1.180	0.961	0.005	8.330

regression coefficient (R^2) imply a variety of adsorption interactions between TBT and the polymers, whereas the low deviation of the n value from 1.0 indicates that minor heterogeneity exists on the surface of the adsorbent [26].

Kinetics of the uptake of TBT by the MIPs (100 ng TBT onto 20 mg of the imprinted polymers) was also investigated, as shown in Fig. 4. The adsorption rate of TBT to MIPs showed two stages, a quick stage at the beginning, and a slow stage before the equilibrium. These results indicated that the imprinted polymers exhibited fast uptake kinetics; 85% of binding was obtained within a short stirring period of 10 min, and adsorption equilibrium was almost reached within 30 min. The RSD values obtained for the MIPs was less than 4.6% at each of these time points (n = 3). The MIPs needed only 30 min to reach adsorption equilibrium for templates, which makes it easy for TBT molecules to reach the imprinting cavities of MIPs. The fast adsorption kinetics (30 min) of the imprinted sorbents is an obvious advantage for the analysis of simple samples.

Three typical OTCs, including the structural analogs MBT, DBT, and TPhT were used to examine the molecular selectivity of TBT-imprinted sites in the synthesized polymers. The amounts of the analytes retained by the imprinted polymers and non-imprinted polymers are shown in Fig. 5. It can be seen that the amount of TBT bound to MIPs is much higher than that bound to the other three OTCs, and the amount of TBT bound to MIPs (Q_{MIP}) is also much higher than that bound to NIPs (Q_{NIP}) (Q_{MIP}/Q_{NIP} = 2.1). This suggests that the template molecule has a relatively higher affinity for the imprinted polymer than its analogs. Because of the steric hindrance effect, TBT fits optimally into the cavities of the imprinted polymers. Moreover, there were no marked differences in the amounts of other organotin molecules (MBT, DBT and TPhT) that bound to MIPs and NIPs, which indicated that non-specific interactions play a significant role in the retention of other OTCs on the imprinted polymer [27]. Therefore, it is suggested that the imprinted sites exhibit a highly selective rebinding of the TBT template molecule to the MIPs.

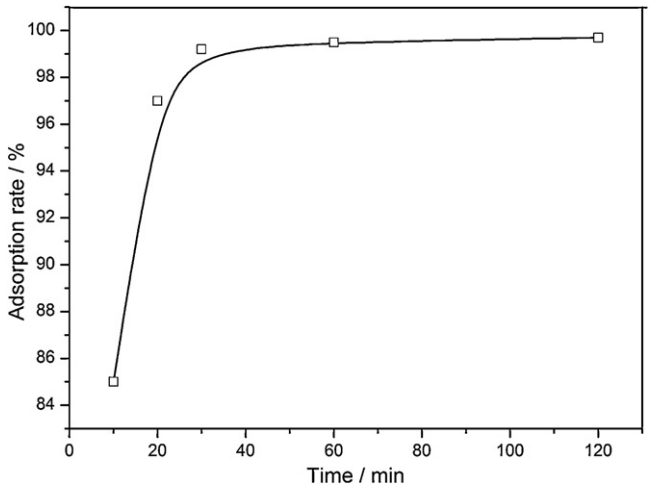

Fig. 4. Kinetic uptake plot of the imprinted polymer.

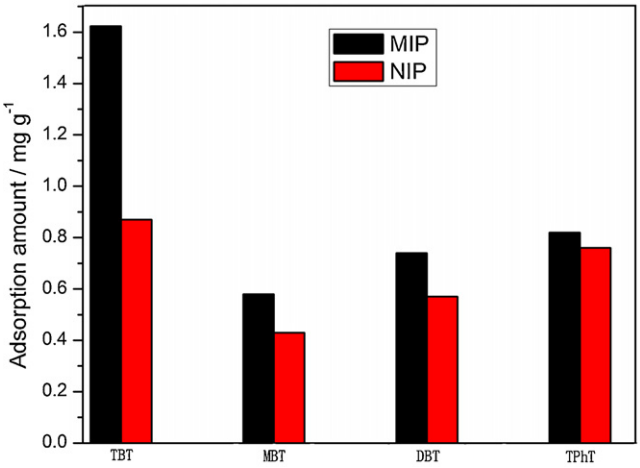

Fig. 5. Adsorption capacities of MIPs and NIPs toward the four organotin compounds in a mixture spiked with TBT, MBT, DBT and TPhT at a concentration of 50 mg/L each.

3.4. Solvent and electrolyte effect on the retention of TBT

Different polar and non-polar solvents (water, acetonitrile, methanol, chloroform, and toluene) were investigated by loading TBT in the range of 10 μg L^{-1}. It was observed that the retention efficiency was higher than 90% for polar solvents, such as water and acetonitrile (Table 2). Retention by the imprinted polymers (M) was not any different than that by the non-imprinted polymer (N) (Table 2, rows 1 and 2), when considering strong non-specific interactions between the polymers and the analytes. However, when electrolytes like Na^+ and Ca^{2+} were added to the loading solution, marked differences appeared between NIP and MIP in the polar solvents, probably because these ions hindered the electrostatic interaction between the positively charged TBT and the acrylamide group (used as a monomer during the polymerization process), resulting in non-specific interactions with the substrate [13].

These experiments demonstrated that TBT has an excellent capacity to be retained on the imprinted polymers in polar solvents even in presence of high concentrations of sodium and calcium ions. Therefore, the method employed here is appropriate to determine the concentration of TBT in seawater or seafood samples.

3.5. Eluting time

Because the template TBT remained in MIPs, it was necessary to elute the MIPs. Typically when the same analyte as the target compound is used as a template in MIP synthesis, the unwashed template molecule may leech during the extraction procedure, leading to false positive results.

MIPs were eluted using the Soxhlet extraction method. The effect of the eluting time was examined by varying the eluting times, 40 mL 0.3 M HCl in methanol (MeOH) as eluent, and it was

Table 2
Effect of solvents and electrolyte addition on the retention of TBT onto the polymers using 20 mg of polymers as sorbents and 10 mL of 10 μg L^{-1} TBT solution in different solvents. N: retention of TBT on the NIP; M: retention of TBT on the MIP.

Retention (%)[a]	H_2O	CH_3CN	CH_3OH	$CHCl_3$	Toluene
(N) TBT	90 ± 3.4	93 ± 2.4	71 ± 4.5	70 ± 4.4	37 ± 3.3
(M) TBT	94 ± 3.3	95 ± 2.5	75 ± 4.6	77 ± 4.7	40 ± 3.6
(N) TBT + 1.0 M Na^+	79 ± 2.8	55 ± 3.6	64 ± 3.7	54 ± 2.4	38 ± 4.4
(M) TBT + 1.0 M Na^+	98 ± 2.5	98 ± 2.6	99 ± 3.3	98 ± 2.4	94 ± 3.5
(N) TBT + 1.0 M Ca^{2+}	58 ± 2.5	40 ± 3.4	49 ± 2.5	99 ± 1.6	98 ± 2.8
(M) TBT + 1.0 M Ca^{2+}	98 ± 2.5	99 ± 1.3	99 ± 2.3	96 ± 1.6	99 ± 1.4

[a] Average ± SD (n = 5).

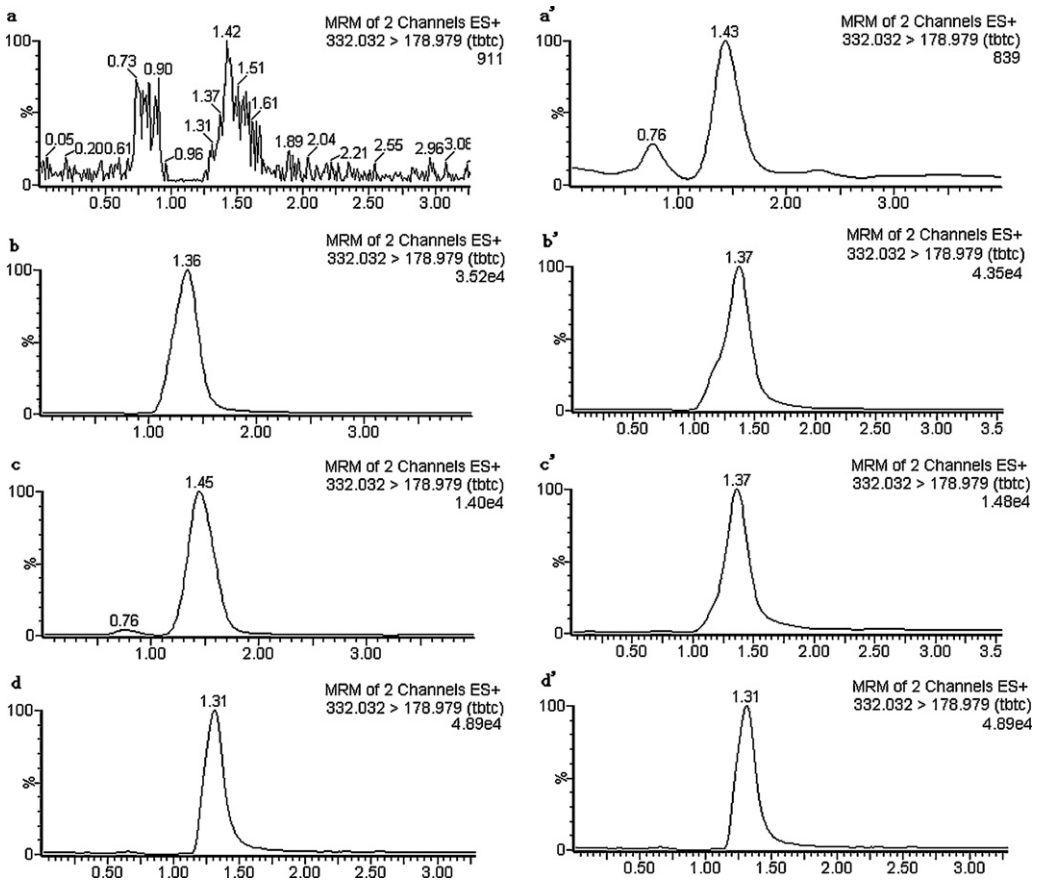

Fig. 6. Determination of TBT in spiked sample solutions using MIP and NIP particles. Chromatograms of 100 μg L^{-1} TBT standard solution, 100 μg L^{-1} TBT spiked solutions of seawater (left), and spiked mussel tissue samples (right): (a), (a′) TBT spiked extraction solution of samples, (b), (b′) spiked sample solution extracted using MIP particles, (c), (c′) spiked sample solution extracted using NIP particles, and (d), (d′) TBT standard solution; TBT injection volume of 10 μL.

detected once every ten hours, and replaced with another the same eluent after detection. It was found that when the eluting times were above six, the S/N has less than 3 dB by LC–MS/MS, which indicated all of template TBT was almost eluted. Therefore, it should be eluted for about 60 h.

3.6. Elution of TBT

An appropriate eluent should be selected to ensure the efficient elution of the analyte from the MIPs and NIPs. Different solvents were compared for their capacity to elute TBT from the imprinted polymers after the washing step, which was introduced to eliminate non-specific binding. Six milliliters of 4:1 MeOH:CH_2Cl_2 [28] were used for washing. TBT could not be eluted with the pure polar and non-polar solvents, such as water, methanol, acetonitrile, and toluene. Some solvent-acidic or solvent-alkaline solutions were also investigated. It was observed that solvent-acidic solutions were more suitable to be used as elution solutions. Moreover, quantitative elution was obtained with 0.1 M HAc in methanol and 0.1 M formic acid in methanol. The latter was found to be more suitable for extraction due to a more reasonable recovery of TBT and better convenience in handling. Therefore, 0.1 M formic acid in methanol was selected as the elution solution in subsequent experiments. We also washed the NIPs with acidic solution, and found that acidic solution has a general effect of washing, because it is a nonspecific adsorption between NIP and TBT, and it can be washed down with organic solvent directly; while the adsorption of TBT by MIP was a specific adsorption with relatively strong binding, so it was difficult to wash down with the organic solvent.

3.7. Analytical characteristics

Quantification of TBT was based on peak area measurements, and external calibration was applied. Limit of detection (LOD) is defined as the minimum concentration of the analyte that can be confidently measured by the method applied. The conventional measurement of the signal-to-noise ratio (3:1) was adopted in our study. According to this method, the LOD of TBT was 0.05 μg L^{-1} (n = 5). The peak area precision (RSD) for five replicate detections of 0.2 μg L^{-1} TBT was 3.8%, with a linear range from 0.1 μg L^{-1} to 20 μg L^{-1} and 5 ng g^{-1}–600 ng g^{-1} in seawater and mussel tissue, respectively. The correlation coefficients (r) were not less than 0.999, and the relative standard deviations were not more than 5%.

3.8. Application of MIP to seawater and mussel tissue samples

To evaluate the usefulness and merit of the imprinted polymer used in this method, seawater and mussel tissue samples (in which no TBT was found and which were treated as described) and samples or extracted samples were spiked with three concentrations of TBT, namely 5, 10, and 50 ng (50, 100, and 500 μg L^{-1}). It required 24 h to extract the sample in order to extract TBT completely for the samples. Because matrix of actual samples is very complex, and it will extend the extraction time. We tested many mussel samples, and found that extraction time of 24 h can be sure to enrich TBT in MIPs completely. The results of TBT extraction using MIPs and NIPs were then compared. As can be observed in Fig. 6, the signal intensity of TBT in the two spiked samples is greatly enhanced when MIP beads are used for TBT extraction (Fig. 6b and b′). Furthermore, the

Table 3
Recoveries of TBT in 50 mL of seawater and 0.1 g of mussel tissue by MIPs ($n = 5$).

Sample	Spiked level	Recovery (%)	RSD (%)
Seawater	0.1 μg L^{-1}	72.7	2.5
	0.2 μg L^{-1}	81.1	2.6
	1.0 μg L^{-1}	67.2	3.7
Mussel tissue	50 ng g^{-1}	94.2	1.3
	100 ng g^{-1}	82.5	4.8
	500 ng g^{-1}	75.0	4.6

sensitivity of this procedure was greater than that of TBT extraction using direct injection analysis (Fig. 6a and a′) or NIP beads (Fig. 6c and c′), likely owing to the stronger interaction of the MIP with TBT. It was obvious that the chromatogram obtained from the MIP bead extraction method was as clean as that shown in Fig. 6d and d′ for the standard solution, which proved that the selectivity of the MIP extraction method was satisfactory.

The TBT recoveries from the seawater and mussel tissue samples by MIPs were 67.2%–81.1% and 75.0%–94.2%, respectively, and the corresponding RSDs were 2.5%–3.7% and 1.3%–4.8%, respectively (Table 3).

The analysis of seawater as well as mussel tissue samples revealed that the presence of other ionic species contained in these samples (e.g., Cu, Pb, Cr, Mn, etc.) had no influence on the retention of TBT onto the polymers. The method is thus interference-free for the environmental and biota samples tested.

4. Conclusions

In this study, a simple MIP was synthesized from complex materials for efficient preconcentration of TBT. It showed a number of advantages, such as fast adsorption dynamics, a high retention capacity and selectivity for TBT, a low detection limit, and great cost-effectiveness (a low price [13]). This polymer may be used for the preconcentration, removal, and determination of TBT in complex environmental as well as biological samples. Thus, a new analytical approach has been achieved in our laboratory by coupling this polymer to LC–MS/MS.

Acknowledgements

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