Research Article

Speciation analysis of mercury by dispersive solid-phase extraction coupled with capillary electrophoresis

A pretreatment method of dispersive solid-phase extraction (DSPE) along with back-extraction followed by CE-UV detector was developed for the determination of mercury species in water samples. Sulfhydryl-functionalized SiO2 microspheres (SiO2−SH) were synthesized and used as DSPE adsorbents for selective extraction and enrichment of three organic mercury species namely ethylmercury (EtHg), methylmercury (MeHg), and phenylmercury (PhHg), along with L-cysteine (L-cys) containing hydrochloric acid as back-extraction solvent. Several main extraction parameters were systematically investigated including sample pH, amount of adsorbent, extraction and back-extraction time, volume of eluent, and concentration of hydrochloric acid. Under optimal conditions, good linearity was achieved with correlation coefficients over 0.9990, in the range of 4−200 μg/L for EtHg, and 2−200 μg/L for MeHg and PhHg. The LODs were obtained of 1.07, 0.34, and 0.24 μg/L for EtHg, MeHg, and PhHg, respectively, as well as the LOQs were 3.57, 1.13, and 0.79 μg/L, respectively, with enrichment factors ranging from 109 to 184. Recoveries were attained with tap and lake water samples in a range of 62.3−107.2%, with relative standard deviations of 3.5−10.1%. The results proved that the method of SiO2−SH based DSPE coupled with CE-UV was a simple, rapid, cost-effective, and eco-friendly alternative for the determination of mercury species in water samples.

Keywords:
Capillary electrophoresis / Dispersive solid-phase extraction / Mercury species / Speciation analysis / UV detection

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1 Introduction

The pollution of heavy metals has drawn widespread attention owing to their high toxicological effects and bioaccumulation properties. As one kind of common heavy metal in environment, mercury (Hg) has different chemical existence forms, showing huge gap in the toxicity to human [1, 2]. The organic forms of mercury such as methylmercury (MeHg), ethylmercury (EtHg), and phenylmercury (PhHg) have much more fatal impact than an inorganic mercury because of the lipophilicity and teratogenicity of organic ones [1−4]. Once entering the organism, the organic mercury is almost impossibly discharged from body. So, recognition and detection of the organic Hg is quite significant rather than determination of the total Hg.

Chromatographic technologies currently available for separation and determination of mercury species generally include GC [4, 5], HPLC [6, 7], and CE [8−10]. The separation technologies are usually coupled with different types of detectors such as ICP-MS [11−13], UV [14, 15], and atomic fluorescence spectrometry (AFS) [16, 17]. In recent decades, CE has boomed rapidly in element speciation analysis [10,18], mainly attributed to its prominent advantages such as high separation efficiency, low sample/solvent consumption, varied separation modes, and short-analysis time [10, 18−22]. Due to the simplicity and low-cost of instrumentation, a UV absorbance detector is the most common detector integrated to commercial CE instruments. However, CE-UV suffers from a primary deficiency of low sensitivity, as the

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Abbreviations: AFS, atomic fluorescence spectrometry; DLLME, dispersive liquid-liquid microextraction; DSPE, dispersive solid-phase extraction; EF, enrichment factor; LLLME, liquid−liquid−liquid microextraction; MPS, 3-mercaptopropyltrimethoxysilane; TEOS, tetraethyl orthosilicate

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result of a low injection volume and narrow optical path-length [21]. Two basic approaches can be adopted to resolve the sensitivity problem, either with special detectors that are more sensitive than UV such as ICP-MS [10], or with increased analyte amounts by using on column and/or off column preconcentration methods [23]. Consequently, developing high-efficiency enrichment techniques and methods for improvement in sensitivity can allow a wider application of CE-UV and avoid the use of complex and bulky instrumentation.

On the other hand, sample matrices in environment are usually complicated and contents of mercury species are usually at trace levels, which would render direct analysis quite difficult or scarcely possible. Direct injection of crude samples/extracts is usually not recommended, since complicated matrices can inhibit or enhance analytes’ ionization or seriously pollute a separator/detector, and concentrations of analytes are too low to detect [23, 24]. Accordingly, effective sample preparation/pretreatment steps before instrumental analysis are critical, aiming at elimination of matrix interfering agents and analyte preconcentration. For this regard, a number of pretreatment technologies/methods for the analysis of mercury species have been developed [10], including SPE [25, 26], stir bar sorptive extraction (SBSE) [5], dispersive liquid-liquid microextraction (DLLME) [14, 18, 27], liquid–liquid-liquid microextraction (LLLME) [9, 15], single drop microextraction [28], dual-liquid point extraction [29], and so on. Dispersive SPE (DSPE) has recently gained popularity [30], in which adsorbents are directly added into sample matrices without packing in a column and conditioning, and then analytes are flushed off by elution solvent and finally the elution solvent is reserved and analyzed. DSPE can avoid tedious treatment in conventional SPE procedure, easy blockage of adsorption column, excessive solvent consumption as well as poor repeatability [31]. At the same time, one major drawback of DSPE is lack of selectivity, easily leading to undesirable consequences and lower detection sensitivity. So, selective adsorbents are highly desirable for DSPE [31, 32].

As is well known, Hg species can well interact with sulphydryl groups [3, 10, 18], which can be utilized for preparing selective adsorbents, such as sulphydryl (or mercapto) functionalized silica particles [33–35]. The mercapto-silica-related studies have been carried out and also applied to mercury measurement [36–38]. Therefore, in this work, we synthesized a kind of adsorbent named SiO$_2$–SH by a facile sol-gel process followed by a simple functionalization procedure, based on the fact that mercury and sulphydryl can form stable metal chelate complexes. And then the SiO$_2$–SH microspheres were used as adsorbents of DSPE coupled with CE-UV for the simultaneous separation and determination of three organic mercury species. Several variables affecting DSPE efficiency were investigated in detail, including sample solution pH, adsorbent amount, extraction and back-extraction time, and eluent composition and volume. Under the optimized conditions, the DSPE-CE method was well validated and successfully applied to real water sample analysis, demonstrating high sensitivity, repeatability, simplicity, rapidity, good practical feasibility, and environmental benignity.

2 Materials and methods

2.1 Reagents and samples

The standard solutions of MeHg and PhHg at a concentration of 1000 mg/L prepared by dissolving the appropriate amounts of the standards in MeOH were both purchased from Aladdin (Shanghai, China). EtHg standard solution at 60 mg/L was purchased from National Institute of Metrology (Beijing, China). Working solutions were prepared by stepwise diluting the stock solutions to the required concentrations using ultrapure water and all stored in refrigerator at 4°C before use. Tetraethyl orthosilicate (TEOS), sodium hydroxide (NaOH), and L-cysteine (L-Cys) were all purchased from Sinopharm Chemical Reagent (Shanghai, China). 3-Mercaptopropyltrimethoxysilane (MPS) used as sulphydrylization reagent was purchased from Sigma–Aldrich (Shanghai, China). Analytical grade ethanol and boric acid (H$_3$BO$_3$) were purchased from Tianjin Kernel Chemical Reagent Factory (Tianjin, China). Deionized ultrapure water used throughout the work was produced by a Milli-Q Ultrapure water system (Millipore, Bedford, MA, USA). The pH measurement was performed through a pHs-3TC digital pH meter equipped with a combined glass–calomel electrode (Shanghai, China).

Lake water samples were collected using a teflon bottle from an artificial lake named Sanyuan Lake located in the schoolyard of Yantai University. Tap water samples were collected from our laboratory of Yantai Institute of Coastal Zone Research, after flowing for about 5 min when needed. The collected water samples were only simply filtrated through 0.45 μm membrane (Tianjin Jinteng Experiment Equipment Ltd., Co., Tianjin, China) to remove possible impurity, and then were stored at 4°C in a refrigerator for use.

2.2 Instrumentation and CE conditions

Mercury determination was carried out by using a P/ACE MDQ CE system (Beckman Coulter, Fullerton, CA, USA) equipped with a diode-array detector. Bare fused-silica capillaries were used, which were purchased from Yongnian Photocductive Fiber Factory (Hebei, China), with inner diameter of 75 μm, outer diameter of 375 μm, total length of 50.2 cm, and effective length of 40 cm. New capillaries were initialized with water (10 min), 1.0 mol/L NaOH (40 min), water (10 min), and running buffer (30 min) for the first use. Before daily experiments, the capillary was reconditioned by rinsing with water (2 min), 1.0 mol/L NaOH (10 min), water (5 min), and running buffer (5 min) sequentially. Between two successive CE runs, the capillary was rinsed with the running buffer for 3 min.
2.3 Synthesis of sulphydryl-functionalized SiO2 microspheres (SiO2−SH)

The whole preparation process of sulphydryl-functionalized SiO2 microspheres was schematically shown in Fig. 1. Firstly, SiO2 microspheres were synthesized by a sol–gel process with hydrolysis of TEOS and aqueous ammonia (NH₃·H₂O) used as catalyst, based on the Stöber method [39]. On the whole, ethanol (30 mL), NH₃·H₂O (5 mL), and ultrapure water (50 mL) were added sequentially to a 250 mL three-necked, round-bottom flask. The mixture solution was magnetically stirred for homogeneous dispersion followed by adding TEOS hydroxyl. So, a pH value of 7.0 was chosen, and it was constant. The efficiency decreased obviously as the pH exceeded 7.0, which was probably owing to the interaction between mercury species and mainly existent hydroxyl. Therefore, 5 mg of adsorbent was chosen to be used as back-extraction solvent of L-Cys did not effectively extract the analytes from the adsorbent [40]. Hence, the effect of sample pH on extraction efficiency was investigated. The testing solution pH was adjusted in the range of 5.0–10.0. As can be seen from Fig. 3A, there was a slight increase of extraction efficiency with the increase of pH value from 5.0 to 7.0. The efficiency decreased obviously as the pH exceeded 7.0, which was probably owing to the interaction between mercury species and mainly existent hydroxyl. So, a pH value of 7.0 was chosen, and it was convenient that the tested samples could be directly analyzed without adjusting the pH values in the following work.

The amount of synthesized adsorbent was studied to obtain the optimal extraction efficiency. Different amounts of adsorbent ranging from 5 to 25 mg were used to adsorb mercury species. The results in Fig. 3B showed that 5 mg of adsorbent was sufficient for high extraction efficiency. Further increasing the amount to 25 mg reduced the extraction efficiency, which was chiefly because the back-extraction solvent of L-Cys did not effectively extract the analytes from the adsorbent to the aqueous solutions. Therefore, 5 mg adsorbent was chosen for further studies.

In the present DSPE procedure, extraction time was defined as the time of oscillation or shaking after the formation of suspension solution by ultrasonic. The influence of extraction time was investigated within 0–3 h. According to the results, no significant change occurred as the extraction time increasing. This phenomenon can be attributed to the good dispersibility of SiO2−SH microspheres in aqueous solution.

2.4 DSPE procedure

Aqueous solution (10 mL) with the pH adjusted to 7.0 (by using 1 mol/L HCl or 1 mol/L NaOH) containing three mercury species was placed into a 15 mL centrifuge tube with conical bottom, followed by adding sulphydryl—functionalized SiO2 materials (5 mg). After ultrasound, the mixture solution was shaken for 1 h at 200 rpm. Then, it was centrifuged for 5 min at 6500 rpm. The supernatant liquid was removed. 0.1% w/v L-Cys (20 μL) containing HCl (0.05 mmol/L) used as back-extractant was mixed with the sedimentary phase. After sonification for about 1 min, the mixture was centrifuged for 5 min at 7000 rpm. The supernatant liquid was taken out and prepared for CE analysis. Illustration of the whole DSPE procedure was shown in Fig. 2.
Combining our comprehensive consideration, the extraction time was set at 1 h.

According to our previous work [41], 0.1% w/v L-Cys solution was selected as eluent to back extract the mercury species and form hydrophilic complexes for CE analysis. Then the effect of L-Cys volume was examined in the range of 20–120 μL with 20 μL interval. As shown in Fig. 3C, high extraction efficiency was attained with 20 μL of L-Cys solution as eluent. The extraction efficiency decreased as the further increase of eluent volumes. It indicated 20 μL eluent could effectively extract the mercury species with preferable enrichment factor. The volume less than 20 μL was not studied for the fact that removal of the supernatant aqueous phase became more difficult with less volume of L-Cys solution. Hence, 20 μL of 0.1% w/v L-Cys was selected for back extraction. In addition, the back extraction time was investigated by changing the ultrasonic time from 0.5 to 10 min. There was no significant signal increasing with the time increasing. So, 1 min was chosen for the ultrasound-assisted back extraction.

In the procedure of desorption, the addition of hydrochloric acid in the eluent of L-Cys solution will be conducive to extract the analytes from the adsorbent materials [42]. A series of 0.1% w/v L-Cys solutions containing different concentrations of HCl ranging from 0–0.25 mmol/L were investigated. As shown in Fig. 3D, the optimal extraction efficiency was achieved in the 0.05 mmol/L HCl and the peak area decreased with the concentrations further increasing. So, 0.05 mmol/L of HCl was used for the work.

Furthermore, during the DSPE procedure, the adsorption and extraction mechanism can be briefly deduced as below. Three organic mercury species could well interact with the sulfhydryl of SiO₂–SH microspheres [3, 43]. So, mercury species were adsorbed by the SiO₂–SH adsorbent. Then, L-Cys was added as eluent (to destroy the former interactions) to produce hydrophilic mercury–L-Cys complexes [18, 29] for CE analysis.

### 3.2 Method validation of the DSPE-CE

Under the above-optimized conditions, the three types of mercury species could be resolved well via the proposed DSPE-CE. To validate the method, several main parameters were determined including linear range, LOD, LOQ, enrichment factors (EFs), precision, and anti-interference ability. Related data were summarized in Table 1 and 2. Good linearity between peak areas and concentrations was obtained in the range of 2–200 μg/L for MeHg, PhHg, and 4–200 μg/L for EtHg, respectively, and the correlation coefficients were all over 0.9990 (Table 1). The LODs and LOQs were assessed based on signal-to-noise ratio of 3 and 10, respectively. As listed in Table 1, the LODs were 1.07, 0.34, and 0.24 μg/L for EtHg, MeHg, and PhHg, respectively, as well as the LOQs were 3.57, 1.13, and 0.79 μg/L. The present sensitivity meets trace analysis requirements and can contribute to mercury species detection in water samples especially industrial and sanitary wastewater. The EF values were calculated via the ratio of the analyte concentration in the sedimented phase to the initial aqueous sample concentration, providing 109, 184, and 120 for EtHg, MeHg, and PhHg, respectively, and showing high enrichment ability. Moreover, as seen from Table 2, the RSDs for intraday precisions in terms of migration time and peak area were less than 0.18 and 5.12%,
Figure 3. Effects of pH (A), material dosage (B), L-Cys volume (C), and concentration of hydrochloric acid (D) on DSPE of mercury species. Five parallel experiments were carried out ($n=5$). CE conditions: injection pressure, 0.5 psi for 5 s; separation voltage +20 kV; detection wavelength, 228 nm; running buffer, 10 mM H$_3$BO$_3$, and 10% v/v MeOH adjusted to pH 9.0 with 1 mol/L NaOH.

Table 1. Parameters of analytical performance of the optimized DSPE-CE for mercury species

| Hg species | Slope (RSD, %) | Intercept (RSD, %) | Linear range ($\mu$g/L) | $r$ | LOD ($\mu$g/L) | LOQ ($\mu$g/L) | EF |
|------------|----------------|--------------------|--------------------------|----|--------------|---------------|----|
| EtHg       | 182.27(4.20)   | -868.99(431.63)    | 4–200                    | 0.9992 | 1.07         | 3.57          | 109 |
| MeHg       | 290.62(4.80)   | -281.20(398.59)    | 2–200                    | 0.9994 | 0.34         | 1.13          | 184 |
| PhHg       | 338.46(7.39)   | 256.88(693.60)     | 2–200                    | 0.9990 | 0.24         | 0.79          | 120 |

a) RSD, $n=5$.

Table 2. Precision of migration time and peak area for the determination of organic mercury by DSPE-CE

| Hg species | RSD (%, $n=5$) |
|------------|----------------|
|            | Intraday       | Interday       |
|            | Migration time| Peak area      | Migration time| Peak area    |
| EtHg       | 0.18           | 3.37           | 1.58          | 6.89         |
| MeHg       | 0.14           | 3.29           | 1.83          | 5.59         |
| PhHg       | 0.13           | 5.12           | 1.83          | 8.36         |

respectively. As well as, the RSDs for the interday precisions were in the range of 1.58–1.63% for migration time and 5.59–8.36% for peak area. Consequently, the method was highly sensitive and capable of accurate quantification of organic mercury.

Furthermore, the interference effects of the possibly co-existing ions were examined by using some common alkali and alkaline earth metal ions (Na$^+$, K$^+$, and Mg$^{2+}$) and heavy metal ions (Co$^{2+}$, Pb$^{2+}$, Cd$^{2+}$, Zn$^{2+}$, and Cu$^{2+}$) in water samples. Under the optimal conditions, the assays were performed by spiking appropriate amounts of interfering ions (individual or mixture) to 10 mL solutions containing three types of mercury species at the concentration of 50 $\mu$g/L individual. As shown in Fig. 4, the obtained results (recoveries within 92.3–99.8%) showed that 100 times (5000 $\mu$g/L) higher of Na$^+$, K$^+$, and Mg$^{2+}$, respectively, ten times (500 $\mu$g/L) higher of Co$^{2+}$, Pb$^{2+}$, Cd$^{2+}$, Zn$^{2+}$, and Cu$^{2+}$, respectively, and even their mixture of all the eight ions had no significant interferences on the mercury determination. Therefore, the developed DSPE-CE was highly selective and reliable for the determination of mercury species, indicating great application potentials in real water samples.
Figure 4. Interference effects of possible coexisting ions on the determination of mercury species by DSPE-CE. Experimental conditions: three mercury species, 50 µg/L individual; Na⁺, K⁺, and Mg²⁺, 5000 µg/L individual; Co²⁺, Pb²⁺, Cd²⁺, Zn²⁺, and Cu²⁺, 500 µg/L individual; mixture, Na⁺, K⁺, and Mg²⁺, 5000 µg/L individual, together with Co²⁺, Pb²⁺, Cd²⁺, Zn²⁺, and Cu²⁺, 500 µg/L individual. Blank means no addition of the above-mentioned ions. Five parallel experiments were carried out (n = 5).

3.3 Applications of the validated DSEP-CE to real water samples and method performance comparison

To further evaluate the practical application of the validated method, tap water, and lake water were analyzed under the optimal conditions of DSPE-CE. Their typical electropherograms were shown in Fig. 5. As observed, none of mercury species endogenous were detected in the two kinds of water samples (curve a). For the spiked water samples, three resolved peaks were obviously demonstrated, corresponding to EtHg, MeHg, and PhHg, respectively (curve b), indicating the selective enrichment ability and good cleanup effect of DSPE. Table 4 lists the recoveries of the two water samples by adding three levels of the three organic mercury standards. As shown from the table, recoveries were in a range of 62.3–107.2% with RSDs lower than 10.2%. It could be concluded that the developed DSPE-CE method was practically applicable for the simultaneous enrichment, separation, and determination of the three organic species at trace levels in real water samples.

Method performance of the validated DSPE-CE for mercury speciation analysis was compared especially in LODs and EFs with other reported CE-hyphenated or HPLC-hyphenated methods, as summarized in Table 3 [7, 8, 11, 14, 15, 18, 25, 29, 44]. As can be seen from the table, our DSPE-CE-UV method presents the LODs lower than [29], comparable to [11, 14, 15, 18, 25, 44], or higher than [7, 8], as well as the EFs higher than [29], comparable to [18], or lower than [15, 44]. Meanwhile, other advantages/drawbacks of the different approaches from the literatures can be noticed as: the reported LLME [7, 14, 15, 18, 44], LPE [29], and SPE [25] based methods usually require certain amounts of harmful organic solvents; the method of MAE coupled with CE-ICP-MS offers higher sensitivity but needs longer time (25 min) to obtain baseline separation with the more expensive instrument [8]; LPE coupled with HPLC-ICP-MS method requires amounts of toxic organic reagents and high cost instrument [11]. Excitingly, our present method only needs less 5.5 min to attain baseline separation of the three organic mercury species. More importantly, in our method, organic solvent is almost free since working solutions were prepared by diluting the MeOH stock solution with ultrapure water. This is superior to the abovementioned approaches, considering they all use more or less toxic organic reagents. Therefore, on the whole, our developed SiO₂–SH based DSPE coupled with CE-UV possessed remarkable advantages such as highly selective extraction efficiency, high sensitivity, shorter analysis time, and simplicity, rapidity, cost-saving, and eco-friendliness, as well as great practical feasibility.

Figure 5. Typical electropherograms of the blank (A) and spiked (B) real water samples after DSPE with the concentration of 50 µg/L for EtHg, MeHg, and PhHg. Peak identification: 1, 2, and 3 corresponds to EtHg, MeHg, and PhHg, respectively. The optimal DSPE condition: sample volume, 10 mL; 5 mg adsorbing material; 0.1% w/v L-Cys (20 µL) containing 0.05 mmol/L hydrochloric acid used as eluent. CE conditions were the same as Fig. 3.
Table 3. Comparison of mercury speciation analysis based on methods of CE and HPLC

| Detection technique | Pretreatment technique | LOD (µg/L) | EF a) | Ref |
|---------------------|-----------------------|------------|-------|-----|
| CE-UV               | DSPE                  | 1.07, 0.34, 0.24 | 109, 184, 120 | This work |
| CE-UV               | DLLME                 | 1.62, 1.79, 0.23 | 118, 102, 547 | 18 |
| CE-ICP-MS           | MAE b)                | 0.021 (MeHg), 0.032 (EtHg) | – | 8 |
| CE-UV               | dCPE c)               | 45.2, 47.5, 4.1 | 17, 15, 45 | 29 |
| CE-UV               | LLLME                 | 0.94 (MeHg), 0.43 (PhHg) | 324, 210 | 15 |
| HPLC-ICP-MS         | LPE d)                | 0.8, 0.5, 1.0 | – | 11 |
| HPLC-UV             | HF−LLLME e)           | 0.7, 3.8, 0.3 | 215, 120, 350 | 44 |
| HPLC-AFS            | SPE                   | 1.4, 4.3, 0.8 | – | 25 |
| HPLC-UV             | DLLME                 | 0.96 (MeHg), 1.91 (PhHg) | 114, 106 | 14 |

a) Sequence of the analytes: EtHg, MeHg, PhHg.
b) Microwave-assisted extraction.
c) Dual-cloud point extraction.
d) Liquid phase extraction.
e) Hollow fiber-based liquid–liquid–liquid microextraction.
f) HPLC-vapor generation atomic fluorescence spectrometry.
g) Vortex-assisted liquid–liquid microextraction.

Table 4. Recovery and precision of the developed DSPE-CE method used for real water samples (n = 5)

| Hg species | Added (µg/L) | Tap water | Lake water |
|------------|-------------|-----------|------------|
|            | Found ± SD (µg/L) | Recovery (%) | RSD (%) | Found ± SD (µg/L) | Recovery (%) | RSD (%) |
| EtHg       | 10          | 8.8 ± 0.8 | 88.4 | 9.8 | 7.73 ± 0.8 | 77.3 | 10.1 |
|            | 50          | 42.1 ± 2.5 | 84.1 | 5.9 | 34.8 ± 2.6 | 69.6 | 8.0 |
|            | 100         | 94.3 ± 4.8 | 94.3 | 5.1 | 70.1 ± 3.7 | 70.1 | 5.3 |
| MeHg       | 5           | 4.6 ± 0.3 | 91.8 | 7.0 | 4.1 ± 0.2 | 92.8 | 6.1 |
|            | 50          | 53.6 ± 2.5 | 107.2 | 4.7 | 44.2 ± 2.6 | 88.3 | 5.8 |
|            | 100         | 90.7 ± 5.8 | 90.7 | 6.4 | 88.6 ± 5.9 | 88.6 | 6.7 |
| PhHg       | 5           | 3.2 ± 0.3 | 64.1 | 9.8 | 3.1 ± 0.3 | 62.3 | 8.5 |
|            | 50          | 52.9 ± 3.5 | 105.8 | 6.7 | 42.3 ± 2.7 | 84.5 | 6.3 |
|            | 100         | 100.6 ± 6.3 | 100.6 | 6.3 | 106.5 ± 3.7 | 106.5 | 3.5 |

4 Concluding remarks

In conclusion, a synthetic material of SiO2–SH microsphere was firstly used as DSPE adsorbent for the simultaneous extraction and enrichment of mercury species. The method of DSPE combined with CE-UV enabled good analytical performances with high sensitivity, short separation time, and high EFs, and had been successfully applied to real water samples. Moreover, much less sample and reagent consumption, low operating cost, water-phase extraction system, and moderate CE separation conditions, made our method a valuable alternative to the speciation analysis of mercury. In view of the advantages, further explorations on smartly devising and synthesizing green adsorption/extraction materials for suitable extraction techniques will provide promising perspectives for speciation analysis of heavy metals by using CE. Furthermore, continuous efforts should be devoted into the various combinations of versatile off/online enrichment techniques for speciation analysis in complicated matrices, potentially offering higher CE sensitivity and satisfying green sustainable development.

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