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Simultaneous determination of arsenic and mercury species in rice by ion-pairing reversed phase chromatography with inductively coupled plasma mass spectrometry

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ABSTRACT

An analytical method using reversed phase chromatography–inductively coupled plasma mass spectrometry for arsenic and mercury speciation analysis was described. The effect of ion-pairing reagent on simultaneous separation of four arsenic (arsenite, arsenate, monomethlyarsonate and dimethylarsinate) and three mercury species (inorganic mercury (Hg^{II}), methylmecury and ethylmercury) was investigated. Parameters including concentrations and pH of the mobile phase were optimized. The separation and re-equilibration time was attained within 20 min. Meanwhile, a sequential extraction method for arsenic and mercury in rice was tested. Subsequently, 1% HNO_3 microwave-assisted extraction was chosen. Calibration curves based on peak area measurements were linear with correlation coefficient greater than 0.9958 for each species in the range studied. The detection limits of the species were in the range of 0.84–2.41 μg/L for arsenic and 0.01–0.04 μg/L for mercury, respectively. The proposed method was then successfully applied for the simultaneous determination of arsenic and mercury species in rice flour standard material and two kinds of rice from local markets.

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1. Introduction

Arsenic (As) and mercury (Hg) are well known as toxic elements because of their potential risks to human health. Exposure to As or Hg has been linked to an increased risk of many physiological disorders and various types of cancer (Mir et al., 2007). With the rapid development of modern industry, As and Hg are often present as a result of human activities, such as mining/processing of ore and wood treatment (Afton, Kubachka, Catron, & Caruso, 2008). Since As and Hg can be taken up and accumulated by crops, the safety and quality of agricultural products are subject to serious threats from contaminated sources.

Unfortunately, rice, a staple crop for Chinese people, is very efficient in As and Hg accumulation (Ma, Shen, Wu, Tang, Shen, & Zhao, 2014; Ren, Sun, Wang, Luo, & Ma, 2014; Rothenberg et al., 2011; Sommella et al., 2013). Along with drinking water and seafood, the consumption of rice has become the major contributor to As and Hg, thereby causing potential health risks (Sun, Williams, & Zhu, 2009). Studies have shown a positive correlation between health risks and the total As and Hg concentrations of rice (Fang et al., 2014; Qian et al., 2010). The toxicities of As and Hg depend on their chemical speciation. Generally, inorganic As is more toxic than organic As, and the levels of toxicity of As compounds are as follows: arsenite (As^{III}) > arsenate (As^{V}) > monomethlyarsonate (MMA) > dimethylarsinate (DMA) (Gurkan, Kir, & Altunay, 2015; Moreda-Piñeiro, Moreda-Piñeiro, & Romarís-Hortas, 2011). Interestingly, some organic As species such as arsenobetaine and arsenosugars have been proved to be non-toxic (Gómez-Ariza, Lorenzo, & Garcıa-Barrera, 2004; Nam, Oh, Min, & Lee, 2010). Evidence has shown that the major Hg species generally found in biological samples are either methylmecury (MeHg) or inorganic mercury (Hg^{II}) (Doker & Bosgelmez, 2015; Li et al., 2007). The levels of toxicity of Hg compounds are as follows: MeHg > ethylmercury (EtHg) > Hg^{II}. Therefore, measuring the total As and Hg concentration alone is not enough to assess the hazards of As and Hg.

In recent years, a number of analytical techniques have been widely employed for the speciation analysis of As or Hg, including gas chromatography (GC) or high performance liquid chromatography (HPLC) coupled with element specific techniques such as atomic absorption spectroscopy (AAS), atomic emission

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spectrometry (AES), atomic fluorescence spectroscopy (AFS), and inductively coupled plasma mass spectrometry (ICP-MS) (Do, Robinet, Pradeau, & Guyon, 2001; Pasias, Thomaidis, & Piperaki, 2013; Pelcova, Docekalova, & Kleckerova, 2015; Zmozinski, Llorente-Mirandes, Lopez-Sanchez, and Silva (2015)). HPLC techniques are better suited for the separation of As or Hg due to the relatively wide compatibility of mobile phase composition and the easiness of sample preparation (Lin, Chang, & Jiang, 2008; Liu, Zhang, Hu, & Cheng, 2013). ICP-MS also has the advantages of high sensitivity, wide linearity, low detection limit, and multi-elemental analysis (Iserte, Roig-Navarro, & Hernández, 2004). Therefore, coupling of ICP-MS with HPLC is the most widely used technique for the individual speciation of As or Hg that has been employed successfully on a number of different matrices (Khan et al., 2015; Moreno, Garcia-Barrera, & Gomez-Ariza, 2013; Raber et al., 2012; Souza, Campiglia, & Barbosa, 2013). C_{18} reversed-phase chromatography with the ion-pairing reagent tetrabutylammonium hydroxide (TBAH) has been employed in the separation of four selenium and four As species within 18 min by HPLC coupled with ICP-MS (Afton et al., 2008). Currently, however, a single method for the simultaneous separation of common As and Hg species in one chromatographic run has not yet been reported. Gómez-Ariza et al. (2004) established a method for the simultaneous determination of Hg and As species in natural freshwater by HPLC (hydrides generation and cold vapor) coupled with a home modified AFS and a standard AFS system. However the use of modified AFS makes this method somewhat complex, and it cannot be widely used for other matrices. To the best of our knowledge, no information is available regarding the simultaneous speciation analysis of As and Hg in rice using HPLC–ICP-MS.

The main objective of this work was to develop a method for the simultaneous speciation analysis of As and Hg species using HPLC–ICP-MS. Seven common environmentally and biologically observed As and Hg species standards were baseline separated on a C_{18} chromatography column *via* ion-pairing reversed phase chromatography. To illustrate the potential applicability of the proposed method, we successfully optimized the chromatographic method applied to the certified reference materials (CRMs) NIST 1568a rice flour, GBW 10043 rice flour, and two kinds of rice flour from local markets.

2. Materials and methods

2.1. Reagents and materials

All the solutions were prepared with doubly deionized water (DDW) (18 MΩ cm^{-1}, Millipore-Q American). The following commercial products were used: Nitric acid (65%) was purchased from Merck, methanol (HPLC grade) was obtained from Kermel, ammonium dihydrogen phosphate (GR) was purchased from Simopharm Chemical Reagent Co., Ltd, L-cysteine (Reagent Grade) from Solarbio Science & Technology Co., Ltd, and both ammonium acetate (GR) and tetrabutylammonium hydroxide (TBAH) 40% in water (ion-chromatography grade) from Shanghai Aladdin.

The rice flour CRM is GBW 10043 rice flour from the National Standard Substance Center in Beijing, China. As (1 mg/mL) in 1 mol/L HNO_3 and Hg (1 mg/mL) in 1 mol/L HNO_3 for the quantification of total As and Hg were acquired from Aladdin. Standards used for sample spiking and identification are as follows: arsenic acid solution (AsO_4^{3-}; As^V, 32.4 ± 0.7 μg/g); arsenious acid solution (AsO_3^{3-}; As^{III}, 124.3 ± 2.0 μg/g); monomethylarsonic acid solution (MMA, 46.2 ± 1.5 μg/g); dimethylarsinic acid solution (DMA, 97.4 ± 3.3 μg/g); methylmercury solution (MeHg, 65.5 ± 2.5 μg/g); ethylmercury solution (EtHg, 75.3 ± 2.7 μg/g); mercuric chloride ($HgCl_2$ ⩾ 99.5%). All the standard materials were purchased from the National Standard Substance Center (Beijing, China). All standard solutions were prepared by dissolving compounds in DDW at 100 μg/mL As and 10 μg/mL Hg. Working standard solutions (1–20 ng/mL for As species and 0.1–2 ng/mL for Hg species) were prepared daily by diluting the 100 μg/mL As species and 10 μg/mL Hg species mixed standard stock solutions.

2.2. Instrumentation

A microwave (CEM, USA) was used for digesting and extracting samples. For total As and Hg determination, all measurements were carried out with an 7700× ICP-MS (Aglient USA). An Aglient 1260 HPLC with a reverse-phase C_{18} column (150 mm × 4.6 mm, 5 μm, Agilent Eclipse plus, USA) were used for the separation of As and Hg species. The outlet of the chromatographic column was directly connected to the nebulizer of the ICP-MS with a small piece of perfluoroalkoxy (PFA). The instrumental operating conditions are shown in Table 1.

2.3. Sample collection and preparation

The proposed method was validated using NIST 1568a rice flour, GBW 10043 rice flour and two kinds of rice samples from local markets. Sample A was Hunan-grown rice with a relatively high concentration of As, are determined in on our previous investigation (Fang et al., 2014). Sample B was common rice purchased from Liaoning province. All the rice samples were ground to powder, passed through an 80-mesh sieve, and oven-dried at 60 °C for 5 h. The rice flour samples were packaged in clean plastic bags and placed in a refrigerator at 4 °C until analysis.

2.4. Determination of total As and Hg

The total content of As and Hg in rice flour was determined by ICP-MS. The rice flour samples were digested according to the method described by Fang et al. (2014). Approximately 0.500 g of rice four was weighed into digestion vessels, and then added with 5 mL of HNO_3. After soaking for 1 h, 1 mL of H_2O_2 was added, and then placed in the microwave digester. The temperature was raised first to 120 °C in 5 min, then to 160 °C in 5 min and held for 5 min. Finally the digestion temperature was elevated to 180 °C in 5 min and held for 10 min.

Table 1
Instrumental operating conditions.

ICP-MS parameters						
Power	1550 W					
Plasma Ar flow	15.0 L min^{-1}					
Carrier Ar flow	1.05 L min^{-1}					
Isotopes monitored	^{75}As, ^{200}Hg, ^{201}Hg, ^{202}Hg					
Quadrupole bias	−16.0 V					
Octopole bias	−18.0 V					
Dwell time for each isotope	0.1 s					
HPLC conditions						
Ion-pairing RP-HPLC	Agilent Eclipse plus C_{18} (150 mm × 4.6 mm, 5 μm)					
Mobile phase A	5 mmol/L TBAH, 10 mmol/L $NH_4H_2PO_4$					
Mobile phase B	5% (v/v) methanol, 0.1% (m/v) L-Cysteine, 0.06 mmol/L CH_3COONH_4					
pH	7.1					
Flow rate	1.0 mL min^{-1}					
Injection volume	40 μL					
Gradient program						
Time (min)	0	0.5	1.5	10	12	20
%A	100	100	0	0	100	100
%B	0	0	100	100	0	0

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