Food Chemistry 181 (2015) 347–353

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Analytical Methods

Simultaneous determination of four organotins in food packaging by high-performance liquid chromatography–tandem mass spectrometry

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ARTICLE INFO

Article history:
Received 26 June 2014
Received in revised form 2 February 2015
Accepted 22 February 2015
Available online 27 February 2015

Keywords:
Organotin compounds
Food packaging
Simultaneous determination
LC–MS/MS

ABSTRACT

A simple method based on high-performance liquid chromatography–tandem mass spectrometry (LC–MS/MS) was developed for the simultaneous determination of organotins—tri-n-butyltin chloride (TBT), triphenyltin chloride (TPT), dibutyltin dichloride (DBT), and diphenyltin dichloride (DPT)—in plastic food packaging. Samples were prepared by ultrasonic extraction with dichloromethane, followed by dissolution in acetonitrile containing 0.1% formic acid, and purification by an MCX column. The extracts were analyzed by LC–MS/MS in multiple reaction monitoring and positive modes with a C18 column; elution was carried out with a gradient of 0.1% formic acid and methanol containing 0.1% formic acid. The limits of detection for TBT, TPT, DBT, and DPT were 0.1, 0.6, 0.8, and 0.3 μg kg^{-1}, respectively. The recovery of organotins in spiked samples ranged from 68% to 113% (relative standard deviation: 0.4–4.2%). The proposed method was successfully employed to identify the target analytes in plastic packaging used for milk and cake.

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1. Introduction

Organotin compounds (OTs) are widely used in the transportation, sanitation, agricultural, and food industries; they also find applications in stabilizers, antifouling coatings, pesticides, and germicides (Yi, Leung, Lam, Lee, & Giesy, 2012). However, the widespread application of OTs as biocides in antifouling systems, aquaculture facilities, and agriculture since the 1960s has led to adverse effects on marine and freshwater organisms (Clarke & Smith, 2011). In particular, tri-substituted OTs show the highest toxicity to aquatic life (Qiu, Chan, & Leung, 2011). Previous studies have suggested that OTs might lead to harmful health effects in humans as well, including reproductive and developmental abnormalities, immunosuppression, and some forms of cancer (Antizar-Ladislao, 2008). For example, tri-*n*-butyltin (TBT) and triphenyltin (TPT) can inhibit enzyme activity in ovarian cells at concentrations as low as 2 ng mL^{-1} (Saitoh et al., 2001) and promote the development of prostate cancer cells at 100 nM (Titley-O'Neal, Munkittrick, & MacDonald, 2011; Yamabe, Hoshino, Imura, Suzuki, & Himeno, 2000). In addition, long-term exposure to low OT levels can lead to nuclear anomalies in human red blood cells and DNA mutation (Du, Chadalavada, Chen, & Naidu, 2014; González-Toledo, Compañó, Dolors Prat, & Granados, 2002; Huertas, Morillo, Usero, & Gracia-Manarillo, 2007; Wang et al., 2008). Hence, in September 2008, the International Maritime Organization (IMO) enforced a global ban on the application of OT-based antifouling systems in seagoing vessels (IMO, 2008). OTs have been internationally recognized as persistent toxic substances (PTS). In a recent report published by the Government of the Hong Kong Special Administrative Region, OTs have been highlighted to be among the top seven endocrine-disrupting chemicals of concern in food (Centre for Food Safety [CFS], 2012).

Recent studies have identified OTs in water, crop, and food at concentrations that may be highly toxic to aquatic creatures and carcinogenic to humans (Muenpo, Suwanjarat, & Klepal; 2011; Guðmundsdóttir, Ho, Lam, Svavarsson, & Leung, 2011; Ho & Leung, 2014). Therefore, many countries and organizations have established maximum limit standards for OTs. For example, recent EU directives specify an annual average environmental quality standard (AA-EQS) of 0.0002 μg L^{-1} TBT and a maximum allowable environmental quality standard (MAC-EQS) of 0.0015 μg L^{-1} TBT for all surface waters (Directive, 2000; Directive, 2008). That is, there are very strict requirements for the determination of OT

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http://dx.doi.org/10.1016/j.foodchem.2015.02.115
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levels. However, in Asian countries other than Japan and the Hong Kong Special Administrative Region of China, there is no specific legislation controlling the use of TBT in food. This is probably due, at least in part, to the complex procedures involved in sample preparation, high cost of TBT-sensitive instruments, and trace levels of OT compounds found in seawater, all of which render the analysis of TBT difficult. In the past decades, numerous methods have been developed for the analysis of OT compounds, including gas chromatography–flame photometric detector analysis (GC–FPD), high-performance liquid chromatography (HPLC), atomic absorption spectroscopy (AAS), and inductively coupled plasma-mass spectrometry (ICP-MS) (Camino-Sanchez et al., 2012; David, Jacq, Sandra, Baker, & Klee, 2010; Prieto et al., 2010; Van Hoeck et al., 2009). The main disadvantages of these methods include tedious and time-consuming extraction, and derivatization steps prior to the chromatographic analysis. The use of liquid chromatography with fluorescence detection has also been reported for OT analysis (Carro, González, & Lorenzo, 2013). However, single HPLC is incapable of detecting a large number of OTs, which lack chromophores in their structures. Although ICP-MS is a very sensitive technique, the OT structures cannot be identified and the interference of inorganic tin species cannot be eliminated (Ayanda, Fatoki, Adekola, & Ximba, 2013; Fang, Borggaard, Marcussen, Holm, & Bruun Hansen, 2010; Yamashita, Dozono, Takahashi, & Honda, 2012). Among the detection techniques, tandem mass spectrometry (MS/MS) offers a number of advantages: more selective separation, element specificity, low detection limits, and high sensitivity. However, there few analytical method for identification and quantification of OTs by LC–MS/MS in various samples have been reported.

OTs are well known plasticizers, often used as the main component of plastic products such as rubber, wrapping paper, and food packaging materials. The consequent migration of OTs from the packaging material to the food can pose several health hazards. Variable amounts OTs have been found in foodstuff and beverages, including beans, vegetables, fruits, eggs, milk, meat (Marcic, Lespes, & Potin-Gautier, 2005; Qunfang et al. 2004), and wine (Azenha & Vasconcelos, 2002; Campillo, Viñas, Peñalver, Cacho, & Hernández-Córdoba, 2012). The origin of dietary OTs is considered to be direct or indirect contact between different types of plastic and foodstuff (Rantakokko et al., 2008). Jones-Lepp, Varner, and Hilton (2001) described a new method, in which micro-HPLC–ESI-MS was applied for the detection of dibutyltin (DBT) from water polluted by a potable-water polyvinyl chloride pipe. In the European Union and United States, the use of OTs in plastic food packaging is regulated because of the inherent possibility of these compounds to migrate to the food (Hamasaki, 2013). As the sample preparation procedures for food packaging are more complicated than those used for water samples, a method with higher sensitivity should be developed to detect trace OTs in food packaging. Thus far, an LC–MS/MS method that can simultaneously determine TBT, TPT, DBT, and diphenyltin dichloride (DPT) in food packaging has not been described. Therefore, it is very important to develop a simple, sensitive, selective, and rapid method for the quantitative determination of a large number of OTs in food and in the environment.

The aim of the present study was to evaluate the status of food contamination by OTs and the migratory behavior of these compounds from packaging to foodstuff, and to verify the effectiveness of legal provisions adopted by the Chinese government. In this regard, an accurate, rapid, and highly sensitive analytical method based on HPLC–MS/MS is proposed. Reliable and practical extraction methods for the determination of OTs in complex natural matrices are also of considerable interest. Herein, we developed and optimized liquid–liquid extraction (LLE) combined with solid-phase extraction (SPE) for the extraction of DBT, diphenyltin (DPhT), TBT, and triphenyltin chloride (TPhT) from food packaging samples. In the multiple reaction monitoring (MRM) mode, specific pairs were used as monitoring ions and the background effect was suppressed as compared to that in the selected ion monitoring mode. The parameters for extraction, clean-up, and mass detection were also optimized. Thus, we successfully developed an analytical method that can simultaneously detect the abovementioned four OT compounds in food packaging.

2. Materials and methods

2.1. Chemicals, reagents, and instruments

TBT, TPT, DBT, and DPT were purchased from Sigma–Aldrich (St. Louis, MO, USA). Diphenyltin dichloride was purchased from Acros Organics (USA). Toluene, methanol, chloroform, and methylene chloride were purchased from Beijing Chemical Reagent Co. (China). Deionized water (>18 MΩ cm^{-1} resistance) provided by a MilliQ water (Milli-Q Plus system, Millipore, Bedford, MA, USA) was used for preparing all solutions and standards. All glassware used was rinsed with solvent before use.

2.2. Preparation of standard solutions

Stock solutions of each analyte (1000 μg mL^{-1}) were prepared in methanol and diluted to 5 μg mL^{-1} with methanol prior to storage at −20 °C. Analyte mixtures for working standards and sample fortification were prepared in 0.1% formic acid in water and methanol containing 0.1% formic acid, respectively. All stock solutions and mixtures were stored in the dark at 4 °C and analyzed as soon as possible.

2.3. Sample preparation

Milk or cake packaging was cut into pieces of 0.5 cm × 0.5 cm and 2.0 g were ultrasonically extracted in 10 mL dichloromethane for 25 min. The extractions were evaporated to dryness at 50 °C and residue was re-dissolved in 5 mL of acetonitrile with 0.1% formic acid. After 10 min ultrasonic treatment and concentration to the half volume of the extraction solution, the remaining liquids were purified using a MCX-SPE cartridge as follows: (1) MCX columns were activated sequentially with 2 mL methanol and 3 mL 0.15 mol L^{-1} ammonium hydroxide; (2) 1 ml the extraction liquor was loaded into the column; (3) 2 mL 0.1 mol L^{-1} HCl and 1 mL methanol were sequentially added in the column to wash non-specific adsorbed impurities; (4) the cartridge was dried under vacuum for about 2 min to remove solvent residues. The analytes retained were eluted with 3 mL 0.3 mol L^{-1} ammonium hydroxide in methanol. The eluent was collected and evaporated to dryness under a stream of nitrogen. It was reconstituted to 1.0 mL with mobile phase. Then the solution was filtered through a 0.45 μm microporous membrane of mixed cellulose ester for LC–MS/MS analysis.

2.4. Instrumentation conditions

LC–tandem MS analyses were conducted on a system consisting of an Agilent 1200 chromatograph (Agilent Technologies, Palo Alto, CA, USA) coupled to an API 2000 mass spectrometer (Applied Biosystems, AB Sciex, Foster City, CA, USA) equipped with a Turbo Ion Spray (ESI) interface.

The OTs were separated by using a reversed-phase analytical column (Agilent Poroshell 120 EC-C_{18} column, 100 × 4.6 mm, 2.7 μm). The column temperature was maintained at 30 °C. The mobile phase used was 0.1% formic acid in water (A) and methanol

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