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Analytical Methods

Multiresidue analysis of sulfonamides, quinolones, and tetracyclines in animal tissues by ultra-high performance liquid chromatography–tandem mass spectrometry

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ABSTRACT

A multiresidue method for the efficient identification and quantification of 38 compounds from 3 different classes of antibiotics (tetracyclines, sulfonamides, and quinolones) in animal tissues has been developed. The method optimization involved the selection of extraction solutions, comparison of different solid-phase extraction cartridges and different mobile phases. As a result, the samples were extracted with McIlvaine and phosphate buffers, followed by clean-up step based on solid-phase extraction with Oasis HLB cartridge. All compounds were determined by ultra-high performance liquid chromatography–tandem mass spectrometry, in one single injection with a chromatographic run time of only 9 min. The method efficiency was evaluated in 5 tissues including muscle, liver, and kidney, and the mean recoveries ranged from 54% to 102%, with inter-day relative standard deviation lower than 14%. The limits of quantification were between 0.5 and 10 μg/kg, which were satisfactory to support future surveillance monitoring. The developed method was applied to the analysis of swine liver and chicken samples from local markets, and sulfamethazine was the most commonly detected compound in the animal samples, with the highest residue level of 998 μg/kg.

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1. Introduction

Nowadays, the widespread use of antibiotics in livestock husbandry is clearly inevitable. It has brought many benefits with respect to control and prevention of diseases and growth promotion. On the other hand, there are concerns about the presence of their residues in food of animal origin which may pose a health hazard to the consumers. Strict tolerance levels have been set for these compounds in the form of maximum residue limit (MRL), which are normally in the range of parts-per-billion, aiming to minimize the risks to human health associated with the consumption of residues (Chinese Ministry of Agriculture, 2002; European Commission, 2010). Although monitoring programs have been going on for several decades, there is still a growing pressure from governmental agencies and private companies to improve the analytical performance in antibiotic residue analysis, making it necessary to increase the efficiency and reduce the cost and time of these analyses. The most useful way for determining antibiotic residues in animal-derived food is the application of multi-class methods that allow detecting a wide range of compounds in a single analytical process. But antibiotic multi-class analysis is a challenging task taking into account the widely varying physicochemical properties of the different classes of antibiotic. The difficulty in developing such multi-class antibiotic determination is compounded by the low concentrations of analytes in the animal tissues in additional to the inherent complexity of the matrices because of high protein and fat content. The analytical procedure normally includes sample treatment and instrumental determination. The sample treatment is a crucial step to achieve efficient extraction and cleanup simultaneously for different classes of compounds from animal tissues. The currently observed trend of determination is to employ liquid chromatography hyphenated with tandem mass spectrometry (LC–MS/MS) because LC is effective in separating non-volatile and thermally labile compounds, and in terms of selectivity and sensitivity, MS/MS represents one of the most powerful detection tools.

Sulfonamides (SAs), quinolones (QNs), and tetracyclines (TCs) are effective antibiotics widely used in human and veterinary medicine. Incorrect use of these drugs or insufficient withdrawal time after treatment can possibly lead to the presence of antibiotic residues in food products, which increases the potential risk to

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consumers in terms of carcinogenic character and allergic reactions (Gentili, Perret, & Marchese, 2005; Littlefield, Sheldon, Allen, & Gaylor, 1990), and contributes to the development of bacterial resistance (Van den Bogaard & Stobberingh, 2000). A number of analytical methods have been described for the determination of sulfonamides (Bedendo, Jardim, & Carasek, 2010; Bogialli, Curini, Di Corcia, Nazzari, & Polci, 2003; Cai, Zhang, Pan, Tie, & Ren, 2008; Economou, Petraki, Tsipi, & Botitsi, 2012; Forti & Scortichini, 2009; Gentili et al., 2004; Hoff, Barreto, & Kist, 2009; Shao et al., 2005; Thompson & Noot, 2005; Yu & Hu, 2012), quinolones (Bogialli, D'Ascenzo, Di Corcia, Laganà, & Nicolardi, 2008; Bogialli, D'Ascenzo, Di Corcia, Laganà, & Tramontana, 2009; Herranz, Moreno-Bondi, & Marazuela, 2007; Herrera-Herrera, Hernández-Borgesa, Rodríguez-Delgado, Herrero, & Cifuentes, 2011; Kantiani, Farré, & Barceló, 2011; Karbiwnyk, Carr, Turnipseed, Andersen, & Miller, 2007; Toussaint, Bordin, Janosi, & Rodriguez, 2002; Toussaint, Chedin, Bordin, & Rodriguez, 2005; Van Hoof et al., 2005; Zhang, Ren, & Bao, 2009), tetracyclines (Andersen et al., 2005; Blasco, Di Corcia, & Picó, 2009; Bogialli, Curini, Di Corcia, Laganà, & Rizzuti, 2006; Carrasco-Pancorbo, Casado-Terrones, Segura-Carretero, & Fernández-Gutiérrez, 2008; Castellari, Gratacós-Cubarsí, & García-Regueiro, 2009; Pena, Lino, Alonso, & Barceló, 2007; Spisso et al., 2009) in animal-food products, respectively. Besides these methods with a group of specific compounds can be measured, there are several residual methods designed for the determination of multi-class antibiotics. Granelli and Branzell (2007) described a screening method for determining five classes of antibiotics (tetracyclines, sulfonamides, quinolones, β-lactams, macrolides) in muscle and kidney by LC–MS/MS. Later, the method for muscle tissues was extended to quantification and confirmation (Granelli, Elgerud, Lundström, Ohlsson, & Sjöberg, 2009). Aguilera-Luiz, Vidal, Romero-González, and Frenich (2008) developed an ultra-high-pressure liquid chromatography–tandem mass spectrometry method (UHPLC–MS/MS) to determine 18 veterinary drugs in milk, including quinolones, sulfonamides, macrolides, anthelmintics and one tetracycline. Carretero, Blasco, and Picó (2008) reported the use of pressurized liquid extraction (PLE) coupled to LC–MS/MS for detecting 31 antimicrobials in meat. Similarly, Jiménez, Rubies, Centrich, Companyó, and Guiteras (2011) applied PLE and LC–MS/MS to the analysis of 41 antimicrobial agents belonging to seven classes in eggs. Freitas, Barbosa and Ramos (2014) and Freitas, Barbosa and Ramos (2015) reported the multidetection of antibiotics from 7 different families in muscle and liver by UHPLC–MS/MS. The main disadvantage of these procedures is the narrow range of the analytes selected from each class. The MRL of SAs is 100 μg/kg, and the combined total residues of all substances within the SA group should not exceed 100 μg/kg (European Commission, 2010). Therefore, it is important to cover as many SA antibiotics as possible in the analytical method. The existing multiclass methods contained only limited number of SAs, which might due to the difficulty in effectively separating the targeted compounds.

The aim of this work is to propose a sensitive multi-residue method, fit for routine official controls, for the simultaneous determination of 38 antibiotics (21 sulfonamides, 13 quinolones, and 4 tetracyclines) in animal muscle, liver, and kidney samples. The method developed combines solid-phase extraction (SPE) sample cleanup and effective UHPLC–MS/MS detection. Analytical performance of the proposed method was evaluated through a validation study which involved assessment of parameters including linearity, specificity, recovery, precision, limits of detection (LODs) and quantification (LOQs). Applicability of the method was demonstrated in the analysis of 51 commercial tissue samples for the presence of antibiotic residues.

2. Experimental

2.1. Materials and reagents

HPLC grade methanol, acetonitrile, and ethyl acetate were obtained from Fisher Scientific Inc. (Pittsburgh, PA, USA). HPLC grade formic acid was purchased from Dima Technology Inc. (Muskegon, MI, USA). Citric acid monohydrate ($C_6H_8O_7{\cdot}H_2O$), disodium hydrogen phosphate dodecahydrate ($Na_2HPO_4{\cdot}12H_2O$), sodium dihydrogen phosphate dihydrate ($NaH_2PO_4{\cdot}2H_2O$), disodium ethylenediamine tetraacetate (Na_2EDTA), and sodium hydroxide (NaOH) were purchased from Beijing Chemical Co. (Beijing, China). Hydrochloric acid and ammonia were obtained from Alfa-Aesar (Ward Hill, MA, USA). Water was purified using a Milli-Q Synthesis system from Millipore (Bedford, MA, USA). Oasis HLB (200 mg) extraction cartridges were supplied by Waters (Milford, MA, USA). Syringe filter was purchased from Pall Corporation (Ann Arbor, MI, USA).

The SA standards sulfacetamide (SC), sulfaguanidine (SG), sulfapyridine (SPD), sulfadiazine (SDZ), sulfamethoxazole (SMX), sulfathiazole (STZ), sulfamerazine (SMR), sulfisoxazole (SIX), sulfamoxol (SMX), sulfamethizole (SMT), Sulfabenzamide (SB), sulfisomidine (SIM), sulfamethazine (SMZ), sulfamonomethoxine (SMM), sulfamethoxypyridazine (SMP), sulfameter (SME), sulfachloropyridazine (SCP), sulfaquinoxaline (SQX), sulfadoxine (SDX), sulfadimethoxine (SDM), sulfaphenazole (SPZ), phthalylsulfathiazole (PST) were all purchased from Sigma–Aldrich (St. Louis, MO, USA). The QN standards norfloxacin (NOR), ciprofloxacin (CIP), pefloxacin (PEF), lomefloxacin (LOM), enrofloxacin (ENR), sarafloxacin (SAR), difloxacin (DIF), oxolinic acid (OXO), flumequine (FLU) were provided by China Institute of Veterinary Drug Control (Beijing, China). Danofloxacin (DAN), ofloxacin (OFL), enoxacin (ENO) were from Sigma–Aldrich (St. Louis, MO, USA). Marbofloxacin (MAR) was from Dr. Ehrenstorfer (Augsburg, Germany). The TC standards tetracycline (TC), oxytetracycline (OTC), chlortetracycline (CTC), and doxycycline (DXC) were supplied by National Institute for Food and Drug Control (Beijing, China).

Standard stock solutions (1 mg/mL) of SAs and TCs were prepared by dissolving 10 mg of individual compound in 10 mL of methanol. For the stock solutions of QNs (1 mg/mL), 10 mg of each QN standard was dissolved in 0.03 mol/L sodium hydroxide and diluted to a final volume of 10 mL with methanol. These solutions were stored at −20 °C and were stable for at least 6 months. Mixed working standard solutions were prepared by diluting stock solution with methanol, respectively. These solutions were stored at −20 °C and were stable for 4 weeks.

The McIlvaine buffer solution was prepared by dissolving 12.9 g of $C_6H_8O_7{\cdot}H_2O$, 10.9 g of $Na_2HPO_4{\cdot}12H_2O$, and 37.2 g of Na_2EDTA in a 1000 mL volumetric flask with about 800 mL of water. The buffer was diluted to volume with water after adjusting the pH to 5.0 with 1 mol/L of NaOH. The phosphate buffer solution was prepared by mixing 190 mL of 0.05 mol/L NaH_2PO_4 and 810 mL of 0.05 mol/L Na_2HPO_4. The SPE elution solution was prepared by combining 150 mL of methanol and 150 mL of ethyl acetate.

2.2. Sample preparation

For routine testing samples, a previously homogenized tissue sample (1.00 ± 0.02 g) was weighed into a 50 mL centrifuge tube. For spiked samples, 50 μL of working solutions at each concentration were added to blank samples. McIlvaine buffer solution (8 mL) was added to each tube, and the contents of each tube were vortex-mixed with a multi-position vortexer for 15 min. The tubes were centrifuged (10,000 rpm, 10 min, 2 °C) and the supernate solutions were transferred. Phosphate buffer (8 mL) was added to the pellets

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