

Development of residual oxytetracycline analysis in cinnamon by dispersive solid-phase extraction using HPLC

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Abstract

The residual oxytetracycline analysis in cinnamon by dispersive solid-phase extraction using high-performance liquid chromatography (HPLC) was developed and validated. Oxytetracycline was extracted using McIlvaine buffer containing ethylenediaminetetraacetic acid (EDTA-2Na). The removal of matrix components derived from cinnamon was carried out using hexane distribution, solid-phase column extraction, and dispersive solid-phase extraction with Z-sep (zirconia-coated silica gel). After the removal of the solvent, the extract was dissolved in a potassium phosphate solution and analyzed by HPLC with a photodiode array detector (PDA). An interfering peak was not observed in the cinnamon extract chromatograms, indicating the high selectivity of this method. Method validation was performed by recovery tests at the maximum residue limit (MRL) concentration (0.1 µg/g) to evaluate the trueness, repeatability (RSD_r), and with-in laboratory reproducibility (RSD_{wr}). The validation results exhibited sufficient recovery (range, 79-81%) and precision (RSD_r ≤ 5% and RSD_{wr} ≤ 7%). These values fulfilled the criteria for the validation guidelines for residual pesticide analysis in Japan. The limit of quantification (LOQ) of this method was 0.03 µg/g in Chinese and Ceylon cinnamon. The time required for the preparation of eight test sample solutions was less than four hours. This method can help inspect residual oxytetracycline in cinnamons.

Keywords : cinnamon, oxytetracycline, dispersive solid-phase extraction, residual analysis, single-laboratory validation study

I Introduction

Cinnamon is a spice obtained from the inner bark of several trees in the genus *Cinnamomum*¹⁾. The most essential cinnamons are Chinese cinnamon (from *C. cassia*) and Ceylon cinnamon (from *C. verum*)²⁾. They are principally used in cooking as condiments and flavoring materials. *C. cassia* and *C. verum* are commercially cultivated as rain-fed crops and are hardy plants³⁾.

In agriculture, antibiotics are used to treat bacterial infections in plants⁴⁾. Oxytetracycline (OTC), a bactericide, is used in many countries to prevent and control bacterial infections in plants⁴⁾. Owing to the easy availability and relatively low cost of OTC, farmers have widely used this bactericide. Actually,

bactericides are used to control bacterial wilt in *C. cassia* and *C. verum*³⁾. Therefore, there is a possibility that OTC can be residual in cinnamon. Antibiotic residues in food due to bactericide use are a concern to consumers worldwide because of their potential for toxicity and allergic reactions, as well as the possibility of pathogens becoming resistant to this drug⁵⁾. Therefore, the MRL values of OTC in foods were set following the introduction of the positive list system in Japan⁶⁾. The MRL values of OTC in “other spice categories” including Chinese cinnamon and Ceylon cinnamon are set at 0.1 ppm (Confirmation date: December 20, 2022)^{*1}. To inspect the safety of cinnamons, it is important to develop a quantitative method to determine the amount of OTC residues in cinnamon.

In the official Japanese method for the determination of OTC

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*1 The Japan Food Chemical Research Foundation : Search engine for MRLs
“http://db.ffcr.or.jp/front/pesticide_detail?id=13800” (Confirmation date: December 20, 2022)