Post-print: Constructing a Pesticide Residue Detection Technology Platform via Ternary Fusion of High-Resolution Mass Spectrometry, Internet, and Data Science

Authors: Pang Guofang, Chen Yi, Fan Chunlin, Bai Ruobin, Sun Yuehong, Chang Qiaoying

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Abstract

This study establishes a pesticide residue detection technology platform based on the ternary fusion technology of high-resolution mass spectrometry, internet, and data science (hereinafter referred to as "ternary fusion technology"), comprising network alliance laboratories, a detection results database and five basic databases, a data acquisition system, and an intelligent data analysis system. Network alliance laboratories distributed across the country upload original pesticide residue detection data via a unified template from their client terminals to the acquisition system; the acquisition system performs data acquisition, information supplementation, derivative consolidation, and toxicity analysis, then conducts contamination level determination with reference to multinational pesticide maximum residue limits (MRL) to form result records, which are stored in the detection results database; the intelligent data analysis system reads data according to user-specified conditions, performs various statistical analyses based on statistical analysis models, generates charts, draws comprehensive conclusions, and then returns the analysis results to the client terminals of the network alliance laboratories. This thereby achieves automatic generation of illustrated pesticide residue detection reports within 20-30 minutes, with "one-click download" capability, representing an operational efficiency impossible to achieve with traditional methods. The pesticide residue detection data platform realizes real-time detection of pesticide residues in edible agricultural products and timely data collection, management, and intelligent analysis, automatically generating relevant pesticide residue detection reports within a short timeframe, thereby providing real-time online services for pesticide residue traceability, risk and safety assessment, and scientific management and use of pesticides. This represents an effective and efficient tool for achieving scientific application of

pesticides, safety supervision of pesticide residues in agricultural products, and ensuring public "safety on the tip of the tongue".

Full Text

Abstract

A technical platform for pesticide residue detection has been developed based on the tri-element integrated technology of high-resolution mass spectrometry, internet, and data science (hereinafter referred to as "tri-element integrated technology"). The platform comprises network alliance laboratories, a detection results database, five foundational databases, a data collection system, and an intelligent data analysis system. Network alliance laboratories distributed across the country upload raw pesticide residue detection data from their client terminals to the collection system using a unified template. The collection system then performs data acquisition, information supplementation, derivative consolidation, and toxicity analysis before determining contamination levels with reference to maximum residue limits (MRLs) from multiple countries, forming result records that are stored in the detection results database. The intelligent data analysis system reads the data according to user-defined conditions, conducts various statistical analyses based on statistical models, generates charts, draws comprehensive conclusions, and returns the analysis results to the client terminals of the network alliance laboratories. This enables automatic generation of illustrated pesticide residue detection reports within 20–30 minutes with "one-click download" capability—a working efficiency impossible to achieve with traditional methods. The pesticide residue detection data platform realizes real-time detection of pesticide residues in edible agricultural products and timely collection, management, and intelligent analysis of data, automatically generating relevant pesticide residue detection reports within a short timeframe. It provides real-time online services for traceability, risk assessment, and scientific management of pesticide residues, serving as an effective and rapid tool for scientific pesticide application, safety supervision of pesticide residues in agricultural products, and ensuring public food safety.

Keywords: high-resolution mass spectrometry, pesticide residue data, intelligent analysis, automatic generation of detection reports

Introduction

In 1976, the World Health Organization (WHO), Food and Agriculture Organization (FAO), and United Nations Environment Programme (UNEP) jointly established the Global Environment Monitoring System/Food (GEMS/Food) program to monitor food contamination in member countries, understand contaminant intake levels, protect human health, and promote international trade development [1]. The United States launched its Pesticide Residue Monitoring Program in 1962, and currently maintains three major pesticide residue monitoring systems including the National Residue Program (NRP) and Pesticide

Data Program (PDP) [2]. The European Union initiated pesticide and veterinary drug residue testing in 1971 under Directive 71/118/EEC, and launched the Community Program for Pesticide Residue Monitoring in 1996, forming a two-tier monitoring system at both EU and member state levels [3]. Japan implemented the world' s most stringent pesticide residue monitoring system—the Positive List System—in 2003, stipulating that only agricultural products complying with this system could enter the Japanese market [4]. China formulated the "Monitoring Plan for Residues in Animals and Animal-derived Foods of the People' s Republic of China" in 1999 [5], and in 2010, five ministries jointly issued the "Food Safety Risk Monitoring Management Regulations," proposing the establishment of a national food safety risk monitoring plan [6].

As food safety has gained strategic importance worldwide, pesticide residue limits have become increasingly numerous and stringent, posing greater challenges for monitoring. Maximum residue limits (MRLs) serve not only as food safety standards but also as thresholds for international agricultural trade and key technical measures for protecting food safety. Currently, approximately 2,000 pesticides are commonly used worldwide. The MRLs established by the EU, United States, Japan, and China reach 162,248 items (for 839 pesticides, 2013), 39,147 items (for over 500 pesticides, 2011), 51,600 items (for 823 pesticides, 2013), and 4,140 items (for 433 pesticides, 2016), respectively [7]. Compared with developed countries, Chinese researchers have broad development space in this field.

Our team conducted a study in 2013 analyzing 4,109 papers on pesticide residue detection published in 15 mainstream international journals between 1990 and 2013. We found that chromatography, long the dominant technology, was quietly surpassed by mass spectrometry around 2001, and by 2013, the number of papers on mass spectrometry for pesticide residue detection far exceeded those on chromatography [8]. Gas/liquid chromatography-tandem mass spectrometry multi-residue detection technology has become a hot research field worldwide [9-16]. Particularly, high-resolution mass spectrometry developed in recent years offers significant advantages in pesticide residue analysis due to its capability to screen pesticides by exact mass, attracting considerable attention from researchers [17-20].

Given the highly digitalized, informatized, and automated nature of high-resolution mass spectrometry detection technology, the resulting data exhibit the "4V" characteristics of big data: large volume, variety, high velocity, and low value density [21,22]. This poses tremendous challenges for data collection, processing, storage, and analysis. Therefore, a pesticide residue detection technology platform capable of rapid intelligent analysis for big data collection, transmission, statistics, and analysis urgently needs development.

To date, no such methods or systems have been reported for constructing a pesticide residue detection data platform based on the integration of advanced high-resolution mass spectrometry technology and data science via the internet, enabling timely collection, management, and intelligent analysis of pesticide residue data in edible agricultural products and automatic generation of relevant detection reports within short timeframes to provide real-time online services for traceability, risk assessment, and scientific management of pesticide residues.

Our team has developed a tri-element integrated technology of high-resolution mass spectrometry, internet, and data science, and constructed a pesticide residue detection technology platform based on this technology. We propose a method for automatic generation and one-click rapid download of detection reports. Through year-round cyclic monitoring of 1,200+ commonly used pesticide chemical contaminants in 146 types of fruits and vegetables across 18 categories by alliance laboratories nationwide using high-resolution mass spectrometry technology, and through internet-enabled interconnectivity and data sharing among alliance laboratories, we have achieved intelligent management and analysis of data via the collection and intelligent analysis systems, enabling automatic report generation and one-click download. This method enhances the capability to detect pesticide residues and improves detection efficiency beyond comparison with any traditional pesticide residue detection technology. It also enables early discovery, early warning, and early management of food safety issues related to pesticide residues, promoting proactive food safety supervision and shifting from passive remediation to prevention.

Platform Architecture

The pesticide residue detection technology platform constructed based on tri-element integrated technology [Figure 1: see original paper] primarily comprises three components: (1) numerous network alliance laboratories distributed nationwide; (2) five foundational databases—including detection sub-databases, pesticide information sub-databases, and geographic information sub-databases; and (3) a monitoring data collection system and intelligent analysis system based on a three-tier architecture of browser, data server, and Web server.

The platform operates as follows: (1) Network alliance laboratories nationwide upload pesticide residue detection results from GC-Q-TOF/MS (hereinafter "GC") and LC-Q-TOF/MS (hereinafter "LC") from their client terminals to the collection system via the internet using a unified template. (2) The collection system forms result records by performing data acquisition, information supplementation, derivative consolidation, toxicity analysis, and contamination level determination with reference to MRLs from multiple countries, then stores them in the detection results database. (3) The intelligent analysis system reads data according to user-defined conditions, conducts various statistical analyses based on statistical models, generates charts, draws comprehensive conclusions, and returns the analysis results to client terminals of network alliance laboratories nationwide. Ultimately, it generates an illustrated detection report.

Establishment of Standard Detection Methods

The established standard method for pesticide residue detection is based on a pesticide exact mass database [Figure 2: see original paper], employing LC and GC technologies for non-targeted high-throughput detection of pesticide residues in fruits and vegetables to obtain relevant raw data and detect pesticide residues, with the process shown in [Figure 3: see original paper].

Establishment of Network Alliance Laboratories

Joining the network alliance laboratories requires meeting four conditions: (1) Familiarity with high-resolution mass spectrometry such as GC, LC, or other HRMS detection technologies for pesticide residues; (2) Certain capability in pesticide residue spectrum interpretation; (3) Passing national or international organization proficiency tests for pesticide residue detection with Grade A (required annually) and passing flying sample tests organized by alliance laboratories; (4) Following the "five unifications" protocol to ensure data uniformity, integrity, security, and reliability: unified sampling, unified sample preparation, unified detection methods, unified data upload format, and unified statistical analysis report content. Through standardized operations by alliance laboratories, regular sampling and testing are conducted annually, quarterly, and monthly, enabling year-round cyclic detection that generates and accumulates large volumes of pesticide residue detection data stored in databases.

Construction of Five Foundational Databases

To effectively preserve and utilize pesticide residue detection data, provide standards for contamination level determination, and offer a basis for statistical analysis, we designed and established a pesticide residue detection database comprising five foundational databases: detection results database, multi-country and regional MRL data sub-database, agricultural product information sub-database, pesticide information sub-database, and geographic information sub-database .

Establishment of Data Collection System

The data collection system employs a three-tier "browser/database server/Web server" architecture. The browser tier resides in client terminals of alliance laboratories as the user interface; the Web server tier at the data center accesses databases and executes preprocessing logic; the database server at the data center stores and manages various pesticide residue data.

The data collection system includes four modules: data acquisition, data preprocessing, contamination level determination, and data storage [Figure 4: see original paper]. Each module functions as follows: (1) Data acquisition module: acquires pesticide residue detection results reported by alliance laboratories. (2) Data preprocessing module: processes reported detection data including validation, supplementation, classification, and consolidation of pesticide, geographic, and agricultural product information. (3) Contamination level determination module: determines contamination levels according to MRLs from various countries/regional organizations. (4) Data storage module: stores result records in the detection results database. The collection system enables automatic upload of detection results and contamination level determination, establishing a pesticide residue detection results database.

Establishment of Intelligent Data Analysis System

The pesticide residue detection intelligent analysis system consists of data layer, access layer, business layer, and presentation layer [Figure 5: see original paper].

(1) Data layer: Comprises the pesticide residue detection results database, four foundational data sub-databases, and related files, providing database and file services and enabling interconnection among databases.

(2) Access layer: Accesses data in databases through database access components and provides it to the business layer.

(3) Business layer: Implements multi-dimensional cross-statistical analysis of sampling points, pesticides, and contamination levels based on statistical models. This system also employs a three-tier "browser/Web server/database server" architecture. The intelligent data analysis system includes six modules: Parameter setting module providing user interfaces and channels for parameter configuration; Single-item analysis module completing 20 single-item statistical functions; Comprehensive analysis module performing five comprehensive analyses based on single-item results; Report generation module creating illustrated detection reports; Appendix generation module producing various statistical tables; Early warning module providing alerts based on analysis results.

(4) Presentation layer: Generates various illustrated intelligent analysis reports based on 20 client-defined conditions. The generated statistical analysis reports include statistical description text, statistical chart data tables, and chart rendering.

Application Demonstration

Scope of Sample Types for Detection

The platform' s intelligent analysis system supports online customization, allowing users to independently select and filter statistical data to highlight interesting or critical information, and customize report types and scopes to enhance data presentation and big data analysis capabilities. The automatic report system also supports customized expansion of report structure and content. For analyzing pesticide residues in fruits and vegetables from 31 provincial capitals/municipalities, the platform enables clients to independently select required report types and download analysis reports.

To investigate pesticide residue patterns and characteristics in fruits and vegetables of different origins sold in 31 provincial capitals/municipalities nationwide, the platform utilized 10 network alliance demonstration laboratories across the country. Following the "five unifications" principle, the laboratories detected residues in 18 categories of 146 fruit and vegetable samples collected from 284 districts/counties and 638 sampling points in 31 provincial capitals/municipalities. LC technology tested 12,551 samples (7,561,620 data entries), while GC technology tested 9,817 samples (6,968,862 data entries). Analysis results were uploaded to the collection system via the network using a unified template. Based on the platform' s five foundational databases, the system supplemented pesticide, geographic, and agricultural product classification information; performed derivative consolidation and pesticide toxicity classification; determined contamination levels according to MRLs from various countries/regional organizations; formed result records; and stored them in the detection results database through unified format data upload.

The intelligent analysis system, based on unified statistical analysis condition settings, has established a prototype of a big database for pesticide residues [Figure 6: see original paper], covering 284 districts in 31 provincial capitals/municipalities with over 600 sampling points; 18 categories of 146 fruits and vegetables with over 20,000 batches of samples; over 400 detected pesticides; 13.74 million detection data entries; 140 million pesticide residue mass spectra; and 25 million illustrated detection reports.

Comparison with Traditional Methods

Compared with existing manual reports, reports generated by this platform offer higher accuracy, speed, and multiple judgment standards, with flexible statistical scope and diverse analysis methods. The platform achieves automation in online data collection, result determination, statistical analysis, and report generation, significantly improving analysis depth, precision, and efficiency—unattainable by traditional statistical reporting modes. This statistical analysis software thus holds extremely important practical significance and commercial promotion value.

Current pesticide residue detection technologies in the United States, EU, Japan, and China still rely on traditional physical standards for qualitative identification, representing targeted detection with limited scope. In contrast, our newly developed technology uses electronic standards for reference, representing non-targeted screening with full-spectrum scanning. In principle, its detection scope is unlimited, offering powerful discovery capabilities and distinct advantages. The US EPA employs 20 detection technologies for over 500 pesticides; the EU (29 countries) uses 15 technologies for 996 pesticides [23]; Japan uses 10 technologies for 832 pesticides [24]; and China currently uses 19 technologies for 609 pesticides. The combined GC and LC technologies can simultaneously screen 1,080 pesticide chemical contaminants, placing it at the international forefront.

Results and Discussion

Innovative Breakthroughs in Pesticide Residue Detection Technology

The GC and LC combined technologies developed based on the tri-element integrated platform represent important breakthroughs in the supply-side reform of pesticide residue detection technology using high-resolution mass spectrometry. The platform achieves six unique innovations: (1) It replaces physical standards with electronic standards as references in traditional qualitative identification, enabling a leap from targeted detection to non-targeted screening. (2) The combined detection capability of the two technologies increases by 51.1% (GC: 485 types) and 39.6% (LC: 525 types) compared with single technologies. (3) A single sample preparation enables effective extraction and purification of over 1,000 pesticide residues; the combined technologies apply to 18 categories of 146 fruits and vegetables. (4) Both combined technologies meet the internationally recognized standard requirement of accurate determination at 10 g/kg. (5) The development of intelligent screening software digitizes, informatizes, and electronizes GC/LC detection of over 1,000 pesticide residues. (6) The combined technology saves resources, reduces pollution, and fully meets the technical requirements of green development, environmental friendliness, and clean efficiency.

The combined technology leads similar technologies in the US, EU, and Japan. It enables early discovery, early warning, and early management of food safety issues related to pesticide residues, promoting forward shift in food safety supervision and gradual transition from passive remediation to prevention.

Comprehensive Understanding of Pesticide Residue Status in China's Fruits and Vegetables

Through automated intelligent statistical analysis, typical regular characteristics of pesticide residues in fruits and vegetables sold in 31 provincial capitals/municipalities were discovered:

(1) **Pervasive existence of pesticide residues**: Using LC technology, 174 pesticides were detected in 12,551 fruit and vegetable samples from 635 sampling points in 31 provincial capitals/municipalities, with 5,486 detections and detection rates of 39%–88% across cities. Using GC technology, 329 pesticides were detected in 9,823 samples from 471 sampling points, with 20,412 detections and detection rates of 54%–97% [Figure 7: see original paper].

(2) **Functional classification of detected pesticides**: LC detected 174 pesticides and GC detected 343 pesticides, totaling 517 pesticides with 93 detected by both technologies. The combined technologies revealed that the top three currently used pesticide types in China are insecticides, fungicides, and herbicides, though detection numbers vary by technology, demonstrating strong complementarity [Figure 8: see original paper].

(3) **Chemical composition classification**: Detected pesticides were primarily organonitrogen, organophosphorus, and organochlorine compounds [Figure 9: see original paper].

(4) **Residue level status**: Currently, detected pesticide residues in Chinese fruits and vegetables are mainly at low to medium levels. Residues not exceeding 10 g/kg accounted for 54.1% (LC) and 51.9% (GC) of detections, showing consistent results between technologies [Figure 10: see original paper].

(5) **Pesticide varieties per sample**: LC showed 52.6% of samples had no detection or only one pesticide, while GC showed 50.0% [Figure 11: see original paper], validating detection accuracy.

(6) **Toxicity status**: Detected residues were primarily low-toxicity and medium-toxicity pesticides, accounting for 85.4% and 92.9% of types and detections (LC), and 83.4% and 87.6% (GC). However, highly toxic pesticides accounted for 3.9% (LC) and 7.0% (GC) of detections, and banned pesticides accounted for 3.2% (LC) and 5.4% (GC), warranting special attention [Figure 12: see original paper].

(7) **Basic safety assurance**: Statistical analysis of pesticide residues in fruits and vegetables sold in 31 provincial capitals/municipalities showed qualified rates of 96.5% (LC) and 96.3% (GC) for vegetables, and 98.3% (LC) and 98.7% (GC) for fruits, demonstrating basic safety assurance [Figure 13: see original paper].

Conclusion

The construction of a pesticide residue detection technology platform based on tri-element integrated technology and its automatic report generation method provides an efficient and accurate data analysis platform for pesticide residue analysis and early warning across China. The network alliance laboratories and standard detection methods ensure data uniformity, integrity, accuracy, security, and reliability. The establishment of the alliance laboratory detection results database and four foundational data sub-databases provides standards and scientific basis for pesticide residue data analysis and contamination level determination. The proposed data collection system enables automatic upload of detection results, data preprocessing, and contamination level determination, establishing a pesticide residue detection results database. The proposed intelligent data analysis system achieves interconnected and interoperable multi-dimensional statistical analysis of pesticide residue data, enabling automated generation of illustrated reports.

The platform can automatically generate illustrated detection reports for a province or municipality within 20–30 minutes with "one-click download" —impossible with traditional statistical methods. Compared with manual reports, the generated reports offer higher accuracy, speed, multiple judgment standards, flexible statistical scope, and diverse analysis methods. The automation of online data collection, result determination, statistical analysis, and report generation significantly improves analysis depth, precision, and efficiency, holding extremely important practical significance and commercial promotion value. This research achievement aligns with the national "13th Five-Year Plan" themes of "enhancing agricultural product safety assurance capacity" and "advancing Healthy China construction," and can play an important technical support role in these fields.

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Note: Figure translations are in progress. See original paper for figures.