DETECTION OF PESTICIDE RESIDUES IN WHITE TEA FRESH LEAVES BASED ON HYPERSPECTRAL AND ARTIFICIAL INTELLIGENCE MODELS /

基于高光谱和人工智能模型的白茶鲜叶农药残留检测

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ABSTRACT

The detection of pesticide residues in white tea fresh leaves is an important step to ensure the quality safety of white tea finished products. Traditional detection methods are costly and inefficient to realize the demand for fast, low-cost, and accurate detection of pesticide residues in white tea fresh leaves. In this study, five types of white tea fresh leaf pesticide residue sample data were obtained using hyperspectral imaging technology for the high-frequency detected pesticides Glyphosate and Bifenthrin, and the SVM and 1D-CNN models were established to detect the samples after noise reduction processing and feature band screening methods. The study shows that the 1D-CNN model has better feature extraction ability, in which the SG-CARS-1D-CNN model has the highest detection accuracy, which is 94.62%, 95.12%, 94.35%, 94.95%, and 95.27% for the five type of species samples, respectively. This study provides pesticide residue detection for white tea fresh leaves based on the combination of hyperspectral data and an artificial intelligence model, which provides an intelligent, nondestructive, efficient, and high-precision pesticide residue detection model for white tea fresh leaves.

摘要

白茶鲜叶农药残留检测是保证白茶成品茶质量安全的重要环节。传统检测方法成本高、效率低,为了实现对白 茶鲜叶农药残留的快捷、低成本、准确的检测需求。本研究针对高频次被检出农药草甘膦和联苯菊酯*,*利用高 光谱成像技术获得 *5* 类白茶鲜叶农药残留样本数据,经过降噪处理和特征波段筛选方法,建立支持向量机 (SVM) 和一维卷积神经网络模型(1D-CNN)对样本进行检测。研究表明,1D-CNN 模型具有更好的特征提 取能力,其中 *SG-CARS-1D-CNN* 模型的检测精度最高,对 *5* 类种样本检测精度分别为 *94.62%*、*95.12%*、 *94.35%*、*94.95%*和 *95.27%*。本研究提供了一种基于高光谱数据与人工智能模型相结合的白茶鲜叶农药残留 检测手段,为白茶鲜叶提供一种智能、无损、高效、高精度的农药残留检测模型。

INTRODUCTION

Tea is the second largest consumer beverage in the world, and China's tea production accounts for about 41.6% of the total global tea production, ranking first in the world, it is the largest consumer of tea in the world, as well as the second largest exporter of tea in the world *(Huo et al., 2024)*.

Anji white tea is a specialty industry in Anji County, Huzhou City, Zhejiang Province, and has been selected as a national geographical indication product in China. Anji white tea is rich in tea polyphenols and tea pigments, which can enhance human immunity, and antioxidant, antimicrobial, and anticancer effects, which are beneficial to human health *(Xia et al., 2024)*. According to the latest statistics, the planting area in the region is more than 200,000 mu, with an annual output of nearly 2,000 tons, an output value of nearly 3 billion yuan, and a comprehensive output value of 5 billion yuan *(Mei et al., 2024)*.

Spraying pesticides is an important method of preserving the yield and increasing the yield of white tea. During the planting process of white tea, weeds competing for soil nutrients and insect pests nibbling have become two key factors affecting the yield of white tea. However, to simplify the management of weeds in the field, reduce labor costs, and increase tea production, some tea farmers have used organic pesticides, such as herbicides and insecticides, in the planting cycle of white tea *(Lin et al., 2023)*. Some tea farmers are not trained in scientific management, and there is abuse and misuse of pesticides, which makes the pesticide residues of tea raw materials exceed the standard. Tea can be consumed directly after brewing in boiling water, and long-term consumption of tea with excessive pesticide residues can directly threaten the life and health of tea drinkers *(Ali et al., 2021)*.

As food quality and safety have become more and more important to consumers, countries and regions such as the United States, Japan, Morocco, and the European Union have set strict Maximum Residue Limits (MRLs) for pesticides in tea *(Luo et al., 2023)*. In the past 10 years, more than 20 countries and regions have notified China of nearly 160 pesticide residues exceeding the MRLs in tea exported from China, which resulted in the return of tea exported from China and the denial of entry, causing huge economic losses. In the past decade, China has revised the National food safety Standard - Maximum Residue Limits of Pesticides in Food for five times *(Yu et al., 2024)*. Among them, the number of pesticide control categories for tea cultivation has been increased and the pesticide residue limits in tea have been reduced.

Excessive pesticide residues in tea are not only a problem of improper supervision of laws and regulations, but also a problem of insufficient detection means. After plucking, white tea leaves are purchased and processed by tea factories, and after withering and drying, they can be brought to the market *(Xiang et al., 2023)*. Therefore, efficient pesticide residue testing of white tea fresh leaves can ensure the food safety of finished tea from the processing source, and also reduce the loss of tea processing factories. This shows the importance of accurate, rapid, and nondestructive pesticide residue detection on white tea fresh leaves.

Currently, the most common methods for pesticide residue detection in tea are gas chromatographymass spectrometry *(Saitoshida et al., 2015)*, liquid chromatography-mass spectrometry *(Shizuka et al., 2018; Ma et al., 2018)*, and so on. The detection accuracy of the above means is high, but it requires standardized detection reagents, standardized detection processes, and professional detectors, and also has the limitations of destructive to samples, time-consuming, and high cost, etc. Currently, cumbersome detection means and high detection costs restrict the universality of pesticide residue detection in tea to a certain extent. As a supplement to gas chromatography and mass spectrometry detection technology, a non-contact, nondestructive, non-polluting, efficient, high-precision, intelligent means of pesticide residue detection in white tea is needed.

Spectroscopic analysis has gradually become a hot spot in the field of pesticide residue detection by its excellent performance, and the most widely used techniques are near-infrared spectroscopy *(Arzu et al., 2020)*, Raman spectroscopy *(Mikac et al., 2021)*, and hyperspectral imaging *(Augustin et al., 2023)*, etc. The spectral imaging technique is an emerging nondestructive testing technique with high spectral resolution, rich spectral bands, and integrated mapping. Hyperspectral imaging technology is an emerging non-destructive testing technology with high spectral resolution and rich spectral bands, which can provide rich spectral-spatial information. At present, hyperspectral imaging technology combined with traditional machine learning models has been applied in the detection of pesticide residues in agricultural products such as grains and vegetables. For example, pesticide residues such as Malathion and Chlorantraniliprole were detected on the surface of grain such as sorghum and corn *(Lu et al., 2021; Zhang et al., 2023)*; pesticide residues such as imidacloprid and cypermethrin were detected on the surface of vegetables such as cucumber, broccoli, and spinach *(Lu et al., 2021; Wang et al., 2024)*. However, research on hyperspectral pesticide residue detection models for tea using smarter deep learning models is significantly lagging behind other agricultural products.

In this study, white tea high-frequency detected pesticides as a research object, using hyperspectral imaging technology combined with noise reduction processing, and feature band screening, respectively, to establish machine learning and deep learning white tea fresh leaves pesticide residue detection model. The aim is to obtain a means of pesticide residue detection for white tea fresh leaves based on the combination of hyperspectral data and artificial intelligence modeling and to provide an intelligent, non-destructive, efficient, and high-precision pesticide residue detection method for white tea fresh leaves.

MATERIALS AND METHODS

Reagents and Samples

The fresh leaves of white tea used in this study were picked on the morning of April 8, 2024, in the white tea plantation of Anji County, Huzhou City, Zhejiang Province, China (30°28′,119°,42′E). The tea plantation is a green tea plantation, i.e., no pesticide spraying or chemical fertilizer application. Two hundred white tea fresh leaves were selected as experimental samples.

The pesticides selected for this study were the high-frequency detected pesticides for white tea: the herbicide glyphosate and the insecticide bifenthrin. Glyphosate is one of the most widely used herbicides in the world and is very effective against perennial weeds. Bifenthrin is a new type of pyrethroid insecticide, which can effectively control aphids, mites, leafhoppers, and many other pests on tea trees.

Glyphosate reagent was used as bifenthrin standard (99.5% purity) from China National Pesticide Quality Supervision and Inspection Center (Shenyang); bifenthrin reagent was used as glyphosate standard (99% purity) from Shanghai Amperexperiment Technology Co. Concerning the latest Chinese national standard (Food Safety National Standard Maximum Residue Limits of Pesticides in Food), the maximum residue limits of tea were calculated to be 1MRLs, i.e., 1 mg/kg for glyphosate and 5 mg/kg for bifenthrin were calculated to be 1MRLs.

The two types of reagent standards were diluted to 1MRLs and 2MRLs to obtain four types of dilutions, i.e., the concentrations of glyphosate dilutions were 1 mg/kg and 2 mg/kg, respectively; the concentrations of bifenthrin dilutions were 5 mg/kg and 10 mg/kg, respectively, and the no-residue control was the laboratory grade one water. White tea leaves were immersed in the four types of dilutions, and 40 white tea leaves were immersed in each type of dilution so that the white tea leaves could fully absorb the pesticides, and a control group was formed with 40 white tea leaves immersed in laboratory grade Ⅰ water.

Hyperspectral imaging techniques and sample data acquisition

The hyperspectral instrument is a Pika XC2 type hyperspectral instrument produced by Resonon Corporation of the United States, with a spectral range of 400 nm~1000 nm, a spectral resolution of 1.3 nm, the number of spectral channels is 447 bands, the number of spatial channels is 1600 bands, a scanning mode of linear push-scanning, and a selection of 23 mm lenses from Schneider, and the detailed technical parameters of hyperspectral imager are shown in Table 1.

Table 1

The hyperspectral data acquisition platform is equipped with an alloy acquisition bracket to fix the hyperspectral imager; four halogen lamps are configured below the lens of the hyperspectral imager to simulate a natural light source; and a servomotor controls the linear movement speed of the sample tray and matches the frame rate of the hyperspectral imager. The hyperspectral data acquisition platform is shown in Figure 1.

Fig. 1 - Hyperspectral data acquisition platform

To minimize the impact of ambient light on data quality, all light sources other than the four halogen lamps were turned off, curtains were used to block the external light and the computer screen light source, and the time of the collection experiment was set in the evening of the day when the tea leaves were collected. The hyperspectral data acquisition platform sequentially collected naturally dried white tea leaves after soaking in reagents, and a total of 200 hyperspectral data were collected.

Data preprocessing and regions of interest

A radiometric correction was performed using Spectral Pro software to convert the raw hyperspectral data DN to reflectance, and the radiometric correction equation is shown in Equation 1.

$$
\lambda_t = \frac{D N_t - D N_d}{D N_w - D N_d} \times \lambda_w \tag{1}
$$

where: λ_t is the reflectance of the degraded indicator feature in the original image, DN_t is the pixel luminance value of the degraded indicator feature in the original image, DN_w is the pixel luminance value of the standard whiteboard, and DN_d is the reflectance of the spectrometer under dark current.

The size of the radiometrically corrected image was 1000 lines×1600 samples×462 bands. To improve the efficiency of the subsequent data processing and at the same time to retain the continuity of the spatial information of the hyperspectral image, the bands with obvious noise effects were eliminated, and the consecutive 238 bands from band1-band227 (401.28 nm-701.37 nm) were retained with the Tea fresh leaf image as the center, cropping out the useless blank areas in the image, and finally obtaining 256 lines × 512 samples x 238 bands.

As shown in Fig. 2, five regions of interest (ROI) were selected inside the leaves of fresh tea leaves, and the size of the selected ROIs was 10 lines x 10 samples, totaling 100 pixels, and the average reflectance of the ROIs was calculated as the reflectance of the samples.

Fig. 2 - Selection of regions of interest for pesticide residues in white tea fresh leaves

The 5 classes of hyperspectral images are each selected 20 imaging effect better labeled regions of interest, a total of 100, a total of 500 ROIs are extracted, 80% of which are used as training samples, 20% of which are used as test samples, the distribution of the number of samples is shown in Table 2.

Data Noise Reduction Processing

Hyperspectral raw data provides rich spectral information to provide a database for pesticide residue detection of white tea fresh leaves, but also contains a large amount of noise interference and sample gaps, as shown in Fig. 3(a). In this study, Savitzky-Golay (SG) smoothing and Moving Average (MA) smoothing methods were utilized for data Noise reduction processing.

Savitzky-Golay smoothing removes the interference of high-frequency noise from the data by polynomial least-squares fitting of the data within the moving window by a polynomial, which is set to w=2 in this study, and is shown in Equation 2; Moving Average smoothing calculates the mean fit by weighting the inter-spectral information, which can maximize the retention of the inter-band information of the spectrum while reducing the noise interference, and in this study, w=2 is set, which is calculated as shown in Equation 3 *(Ma et al., 2023)*.

$$
X_k^* = \frac{1}{H} \sum_{i=-w}^{i=w} X_{k+1} h_i
$$
 (2)

where: k is the center point of the window, h_i is the smoothing coefficient, ${\rm H}=\sum_{i=-w}^{l=w}\hbar_{i}$ $\sum_{i=-w}^{i=w}$ λ_i .

$$
X_k^* = \frac{1}{2w+1} \sum_{i=-w}^{i=w} X_{k+1}
$$
 (3)

where: k is the center point of the window.

Fig. 3 - Noise Reduction Processing Spectral Curve *(a) Original Spectral Reflectance; (b) S-G Spectral Reflectance; (c) MA Spectral Reflectance*

As shown in Fig. 3, the spectral curves (Fig. 3-b, c) processed by the above two noise reduction methods have retained the spectral characteristics of the original spectral curves; compared with the original spectral curves (Fig. 3-a), they have become smoother, the noise interference has been significantly reduced, and the spectral difference between 420 nm-475 nm and 560 nm-620 nm has been enlarged, which possesses a good potential for data analysis.

Characteristic Band Screening

Hyperspectral data provides a large number of spectral bands, but too many spectral bands will lead to data redundancy, which is not conducive to further analysis of the data, in this study, Competitive Adaptive Re-weighted Sampling (CARS) and Successive Projections Algorithm (SPA) two Band feature extraction (Bfe) methods will be used for feature band screening to downsize the data *(Wang et al., 2024)*.

CARS is a feature band screening method that combines Monte Carlo Sampling (MCS) and Partial Least Squares (PLS). In this study, the number of MCS is set to 50, cross-validation is performed 5 times, and the best feature band is obtained after 10 repetitions. SPA is a forward feature variable screening method that can effectively eliminate the original redundant information in spectral data. Based on the data processed by the two previous noise reduction methods, the number of feature bands after feature band screening is shown in Table 3, and the distribution of feature bands is shown in Fig. 4.

Table 3

By comparison, it can be seen that the number of feature bands filtered by CARS is significantly larger than that of SPA, and combining the two noise reduction methods in the previous section, the number of bands filtered by MA-CARS is the largest, with 40 bands, and it is concentrated in the bands where the curvature of the spectral curve varies greatly; the number of bands filtered by SG-SPA is the smallest, 19 bands, and it is also distributed in the places where the curvature varies greatly, but it is more uniformly distributed compared

Table 4

with that of CARS. The number of bands screened by SG-SPA is the least, 19, although they are also more distributed at the bands with larger curvature changes, compared with CARS.

Support Vector Machine Model

In this study, the classic model of machine learning-Support Vector Machine (SVM) will be used to detect the pesticide residue samples of white tea fresh leaves. SVM realizes the classification of different categories of samples by constructing the optimal hyperplane, which has the advantages of good small-sample detection ability, excellent generalization ability, avoiding dimensionality catastrophe, etc. The samples after noise reduction and feature band screening and the full band samples after noise reduction are used as input features of the SVM model.

The samples after noise reduction processing and feature band screening in the previous section and the full band samples after noise reduction processing are used as the input features of the SVM model, and the model performance evaluation of the SVM model Test sample detection is carried out by utilizing the precision of confusion matrix calculation (Equation 4).

$$
Accuracy = \frac{Tp + Tn}{Tp + Tn + Fp + Fn} \times 100\%
$$
\n(4)

where: T is the true target category; F is the false target category; p is the true prediction and n is the false prediction.

The detection accuracies of SVM models for different kinds of Test samples are shown in Table 4. It can be seen from the comparison that the detection accuracy of the samples with feature band screening is higher than that of the full band samples; compared with MA, the SG noise reduction process is more favorable to improve the detection accuracy; among them, the SG-SPA-SVM model has the highest detection accuracy, and the detection accuracies of the four kinds of pesticide residue samples are 91.54%, 92.37%, 90.18%, and 91.86%, and the detection accuracies of the no residue samples are 92.83%, which are 3.97%-5.69% higher than that of the full-band samples. The detection accuracy of the SG-SPA-SVM model is 92.83%, which is 3.97%-5.69% higher than that of the full-band samples.

Comprehensively, the SG-SPA-SVM model extracts fewer feature bands, but the band distribution is more reasonable. The method not only reduces redundant information and improves training efficiency, but also has excellent pesticide residue detection capability.

1D-CNN Model

In this study, deep learning methods will be also use to detect pesticide residue samples in white tea fresh leaves. The convolutional neural network has powerful data feature extraction ability and better generalization. In this study, a 1D-CNN model is built based on Pycharm, and the model structure is shown in Figure 5, which mainly consists of one input layer, six convolutional layers, and fully connected layers. The activation function is ReLU, the optimization algorithm is Adam, the initial learning rate is 0.001, and the pooling layer is Maxpool. The main parameters of the 1D-CNN model are shown in Table 5.

Main parameters of the 1D-CNN model

Table 5

Table 6

The samples after noise reduction processing and feature band screening in the previous section and the original samples are used as the input features of the 1D-CNN model respectively, and the obtained detection accuracies for different kinds of samples are shown in Table 6. It can be seen from the comparison that compared with the original samples, the samples with feature band screening have higher detection accuracy, CARS show better performance, and the SG-CARS-1D-CNN model has the highest detection accuracy, which is 94.62%, 95.12%, 94.35%, and 94.95% for four kinds of pesticide residue samples, and 95.27% for the no residue samples.

RESULTS

In this study, the white tea fresh leaf pesticide residue hyperspectral data samples were processed by means of noise reduction processing, feature band screening, etc., and the pesticide residue samples were detected using SVM and 1D-CNN models, respectively, and the detection accuracies of the models are shown in Table 7.

Table 7 shows that the detection accuracy of the SG-SPA-SVM model is the best in the SVM model; the detection accuracy of the SG-CARS-1D-CNN model is the best in the 1D-CNN model; the detection accuracy of the 1D-CNN models is overall higher than that of SVM models; the SG noise reduction processing shows excellent potential in both kind of models.

Regarding the SVM models, feature band screening has a greater impact on the model detection accuracy, and the SG-SPA-SVM model improves the detection accuracy by 3.97%-5.69% compared to the SG-ALL-SVM model. Noise reduction processing has a smaller impact on the model detection accuracy, and the SG-SPA-SVM model improves the detection accuracy by 0.17%-1.41% compared to the MA-SPA-SVM model.

Regarding the 1D-CNN models, the ALL-1D-CNN model has shown promising detection potential; while noise reduction processing and feature band screening can further improve the detection accuracy of the model, the SG-CARS-1D-CNN model improves the accuracy compared to the ALL-1D-CNN model by 1.23% to 2.19%; compared with noise reduction processing, feature band screening can further improve the model detection accuracy.

CONCLUSIONS

In this paper, the white tea fresh leaf pesticide residue hyperspectral data samples were processed by means of noise reduction, feature band screening, etc., and the SVM and 1D-CNN models were utilized to realize the accuracy detection of white tea fresh leaf pesticide residue samples, respectively. The main results of this study are as follows:

1) Samples of pesticide residues in fresh white tea leaves were prepared, and hyperspectral data of the samples were collected; Through the combination of sample data noise reduction, feature band screening, and other means with deep learning, the optimal model for pesticide residue detection in fresh white tea leaves was obtained: The detection accuracy of SG-CARS-1D-CNN model for four pesticide residue samples was 94.62%, 95.12%, 94.35% and 94.95% respectively, and the detection accuracy of no residue samples was 95.27%.

2) By comparing the SVM models, the samples screened by feature bands can significantly improve the pesticide residue detection accuracy of the SVM model, and the noise reduction process can slightly improve the pesticide residue detection accuracy of the SVM model. Among them, the SG-SPA-SVM model has the highest detection accuracy, which is improved by 3.97%-5.69% compared with the SG-ALL-SVM model.

3) The deep learning 1D-CNN model has better feature extraction ability, and the original data without noise reduction processing and feature band screening show good detection accuracy in the ALL-1D-CNN model. Noise reduction processing and feature band screening can slightly improve the detection accuracy of the 1D-CNN model, in which the SG-CARS-1D-CNN model has the highest detection accuracy, with 4.62%, 94.86%, 94.28%, and 94.95% for the four pesticide residue samples, respectively, and 95.27% for the no residue samples, which is much better compared with the ALL-1D-CNN model, the detection accuracy was improved by 1.23%-2.19%.

In this study, the SG-CARS-1D-CNN model obtained has a good detection ability for pesticide residues in white tea fresh leaves, which provides a new method and means for the detection of pesticide residues in white tea fresh leaves based on hyperspectral data. It provides an accurate, rapid, and non-destructive detection idea for white tea fresh leaves pesticide residues, which is an effective supplement to the traditional detection means.

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