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Fast and non-invasive identification of Baijiu based on Tyndall effect and chemometrics

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1. Introduction

ABSTRACT

The value of Baijiu is affected by its flavor, age, and adulteration. Therefore, a simple and rapid identification method is crucial for the market. In this study, we present a rapid, non-intrusive identification technique for Baijiu utilizing the Tyndall effect combined with chemometrics analysis. Our experiment begins illuminating Baijiu with a 405 nm wavelength laser and recording the resulting bright light path due to the Tyndall effect. To further analyze the color and brightness information, Principal Component Analysis (PCA), Linear Discriminant Analysis (LDA), Hierarchical Cluster Analysis (HCA), and Multilayer Perceptron (MLP) were employed. This study establishes correlations between the brightness of the Tyndall light path and seven trace flavor compounds in Baijiu. The findings demonstrate that this method effectively identifies the flavor, age cellar, and adulteration of Baijiu and also quantitatively detects the concentrations of flavor compounds. Additionally, an analysis platform was developed to enable the rapid identification of Baijiu.

Baijiu, also known as Chinese liquor, is widely popular for its unique flavor. There are more than 2000 trace flavor compounds in Baijiu, such as alcohols, esters, acids, and aldehydes. These compounds account for only about 2% of the total composition, but they are crucial in determining the unique flavor of Baijiu (Dong et al., 2024; Xie et al., 2022; Zheng et al., 2021). In addition, the aging process of Baijiu involves a series of reactions such as volatilization, oxidation, complexation, esterification, hydrolysis, and colloidal molecule formation. These reactions can enhance the flavor and texture of the Baijiu. Consequently, Baijiu stored for a long time will have better taste and higher value (Deng et al., 2020; Jia, Ma, Hu, & Mo, 2023; Zheng et al., 2021).

In the Baijiu market, the prices of different flavors and aging times

vary significantly. Adulterating high-value Baijiu with low-value varieties disrupts the market and harms consumers. However, it is challenging for consumers to distinguish the quality of Baijiu despite many identification methods for Baijiu have been studied (Jia et al., 2023), including sensory evaluation and instrumental analysis.

Sensory evaluation, the most common identification method, relies on tasters to assess the flavors and age of Baijiu (Li, Xu, Yu, & Zheng, 2023; Wu et al., 2023). Thus, this approach is subjective and depends heavily on the tasters' experience.

Instrumental analysis methods can be divided into two categories: quantitative and qualitative. Quantitative analysis methods include chromatography (Li et al., 2023; Wang et al., 2023; Wei et al., 2023), mass spectrometry (Ge et al., 2023; He, Yangming, Gorska-Horczyczak, Wierzbicka, & Jelen, 2021; Sun et al., 2021), and gas

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chromatography–mass spectrometry (Niu, Yang, Mao, & Xiao, 2024; Wu et al., 2023; Zhang et al., 2021). These methods can offer highly accurate quantitative analysis of baijiu. However, they are usually expensive, time-consuming, and require consumable materials like analytical columns. Qualitative analysis methods, on the other hand, include spectrometry (Liu et al., 2022; Tao et al., 2023; Wu et al., 2023; Zong, Zhou, Wen, Gan, & Li, 2024), colorimetry (Jia et al., 2024; Sun, Qian, Zheng, Li, & Lin, 2020; Wu et al., 2022), and electronic nose (He et al., 2021; Wang, Meng, Jin, & Sun, 2021; Zhang et al., 2021). These methods can reduce identification costs but still require significant time and skilled technicians. In addition, some of these methods still require supplementary reagents, increasing both the cost and complexity. Therefore, it is crucial to have a fast and non-invasive method for Baijiu identification.

The Tyndall effect, a phenomenon observed when light passes through a colloid, reveals the light path and has become an effective tool in environmental and biomedical fields due to its high sensitivity and low cost. In environmental pollution studies, the Tyndall effect has been used to detect the concentration of Hg^{2+} by using silver nanoparticles or gold nanoparticles (Deng et al., 2021; Huang et al., 2021). In the pharmaceutical field, ascorbic acid and glutathione concentrations were successfully detected using the Tyndall effect (Gao et al., 2021; Sun et al., 2022). Additionally, the concentrations of three fluorinated pesticides in tea were successfully detected using the Tyndall effect of Ltryptophan nanoparticles (Wang et al., 2023). However, these detections are invasive and will irreversibly contaminate the sample.

In our previous work, we qualitatively studied the Tyndall effect in sauce-flavored Baijiu and found that blue-violet light is the best illumination source to enhance the Tyndall effect (Liu et al., 2021). However, there remains a need for more accurate and non-invasive methods to identify flavor, age, and adulteration in Baijiu.

This work introduces a method combining the Tyndall effect and chemometrics to identify Baijiu. Chemometrics, which is widely used in Baijiu detection, utilizes mathematical, statistical, and computational techniques to process and interpret measurement data (Chen et al., 2023; Ding et al., 2023; Song et al., 2021). The characteristic information of the Tyndall effect, extracted from the captured images, is used to identify the flavor type, cellar age, and adulteration of Baijiu (Scheme 1). The information includes the light path's color coordinate value and the light path's average gray value. These values are analyzed using Principal Component Analysis (PCA), Linear Discriminant Analysis (LDA), Hierarchical Cluster Analysis (HCA), and Multilayer Perceptron (MLP) neural network model to discriminate the flavor type, cellar age, and adulteration of Baijiu. In addition, relationships between grayscale values and concentrations of seven trace flavor compounds are established to detect the concentration of trace flavor substances.

Furthermore, an analysis platform has been developed to improve the data collection efficiency and user convenience. To our knowledge, this is the first report on identifying Baijiu flavor, cellar age, and adulteration based on the Tyndall effect and chemometrics. The presented method offers several advantages: it is fast, simple, non-invasive, lowcost, and highly precise, making it highly promising for Baijiu identification.

2. Materials and methods

2.1. Reagents and materials

Ethanol, acetic acid, lactic acid, caproic acid, acetaldehyde, and acetal were procured from Shanghai Titan Technology Co., Ltd. (Shanghai, China). Ethyl acetate, ethyl hexanoate, and isoamyl alcohol were obtained from Shanghai Boer Chemical Reagents Co., Ltd. (Shanghai, China). Ethyl lactate was sourced from Shanghai Mayer Biotech Co., Ltd. (Shanghai, China). Purified water was purchased from Hangzhou Wahaha Group Co., Ltd. (Hangzhou, China).

The Baijiu sample "Dongfang Mingzhu Laojiang 15 Baijiu" was acquired from Hubei Dongfang Mingzhu Liquor Co., Ltd. (Xiangyang, China). Other Baijiu samples, including those of strong flavor, light flavor, rice flavor, fuyu flavor, feng flavor, herbal flavor, soybean flavor, sesame flavor, te flavor, mixed flavor, laobaigan flavor, and sauce flavor types, as well as sauce flavor Baijiu aged for 5, 10, and 15 years, were all purchased from Jiangsu Guanyun Liquor Co., Ltd. (Suqian, China). The information of all the samples is shown in Table 1. These samples encompass a diverse range of Baijiu categories, and thus providing a comprehensive selection for the study.

Table 1

Details of Baijiu samples	s.
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Baijiu	Flavor	Cellar Age	Abbreviation
Strong Flavor Baijiu	Strong	-	F1
Light Flavor Baijiu	Light	-	F2
Rice Flavor Baijiu	Rice	-	F3
Fuyu Flavor Baijiu	Fuyu	-	F4
Feng Flavor Baijiu	Feng	-	F5
Herbal Flavor Baijiu	Herbal	-	F6
Soybean Flavor Baijiu	Soybean	-	F7
Sesame Flavor Baijiu	Sesame	-	F8
Te Flavor Baijiu	Те	-	F9
Mixed Flavor Baijiu	Mixed	-	F10
Laobaigan Flavor Baijiu	Laobaigan	-	F11
Sauce Flavor Baijiu		-	F12
Sauce Flavor Baijiu aged 5 years		5 years	Y5
Sauce Flavor Baijiu aged 10 years	Sauce	10 years	Y10
Sauce Flavor Baijiu aged 15 years		15 years	Y15
Dongfang Mingzhu Laojiang 15 Baijiu		Mix	DF

Note: The symbol "–" represents Baijiu not aged in cellars or for less than one year. "Mix" denotes a blend concocted from Baijiu of various cellar ages, ranging from 3 to 25 years.



Scheme 1. Fast and Non-invasive Identification of Baijiu Based on Tyndall Effect and Chemometrics.

2.2. Preparation of trace flavor compound solutions

To prepare 500 mL solution of 53% (ν/ν) ethanol, 256 mL of ethanol is mixed uniformly with purified water. 50 mL solution of 1000 mM ethyl lactate was then prepared by uniformly mixing 0.05 mol of ethyl lactate with the 53% (ν/ν) ethanol solution.

For the preparation of eight bottles containing 20 mL each of ethyl lactate solutions with concentrations of 500, 200, 100, 50, 20, 10, 5, and 1 mM, the following volumes of the 1000 mM ethyl lactate solution are used: 10000, 4000, 2000, 1000, 400, 200, 100, and 20 μ L, respectively. Each volume is uniformly mixed with 53% (v/v) ethanol solution.

Using the same method, solutions of acetic acid, lactic acid, caproic acid, acetaldehyde, acetal, ethyl acetate, ethyl hexanoate, and isoamyl alcohol with concentrations ranging from 1000 to 1 mM are prepared.

In all the identification experiments conducted in this study, the concentration of each sample is maintained consistently without dilution. The 53% (v/v) ethanol solution prepared from ethanol and purified water is used as a blank sample.

2.3. Measurement of emission spectra

The emission spectra are measured using an FS5 Spectrofluorometer (Edinburgh Instruments, UK). The specific operation of this instrument is as follows:

- 1. Clean cuvettes containing Baijiu samples are securely placed in the fluorometer's holder.
- 2. The excitation wavelength is set to 405 nm, same as the wavelength of the laser used, with an excitation slit width of 3 nm.
- 3. The emission wavelength range is set from 425 to 750 nm, within visible light range with an emission slit width of 5 nm.
- 4. The scanning interval is set to 1 nm, and emission spectrum data for each sample were measured and recorded.

2.4. Image acquisition

The Image acquisition setup, depicted in Fig. S1, consists of a blueviolet laser, Baijiu samples, a square quartz sample cell, and a camera. The laser operates at a wavelength of 405 nm with a power output of 10 mW. The quartz sample cell measures $30 \times 30 \times 30$ mm, with transparent walls and a frosted bottom. The camera is equipped with a zoom lens (Computar, Japan) and captures images at a resolution of 1280 × 1024 pixels.

During image collection, the laser is fixed to the side of the sample cell, and the direction of the laser beam is directed perpendicular to the cell wall. The Baijiu sample is then illuminated by the laser with a spot diameter of 3 mm, and create light columns that are perpendicular to the light path.

Prior to image capture, both the laser and camera are preheated to ensure stability. The procedure involves the following steps:

- 1. Pour 15–20 mL of the Baijiu sample into a clean sample cell, and make sure the liquid level is higher than the laser beam.
- 2. Place the cell in the fixture of the acquisition device, and ensure the walls are clean and the shooting is unobstructed.
- 3. Turn off all light sources except the laser in the darkroom anechoic chamber to capture the image.

Each sample is imaged six times. The inner walls of the cell are replaced or cleaned before each new sample to prevent crosscontamination. It is worth noting that the Baijiu samples remain uncontaminated and can be almost completely recycled as long as the sample container is aseptic and hygienic.

2.5. Data analysis

Before analysis, it is necessary to extract image information, including color data and grayscale data, through the following steps:

- 1. Image Preprocessing: The edges of the image are cropped, keeping only the area showing the Tyndall effect. The cropped image is resized to 660×660 pixels.
- 2. Noise Removal: Background noise, including darker noise, stray light, and bright noise, is eliminated. Threshold segmentation is used to remove darker noise by selecting all bright areas. Stray light and bright noise are removed using an opening operation involving erosion followed by dilation.
- 3. Data Extraction: color data and grayscale data are extracted. For the color data, the x and y coordinates in Commission Internationale de L'Eclairage (CIE) chromatic diagram are obtained by averaging the RGB values of all non-black pixels. In order to extract gray value, a region of 120×660 pixels at the center of the image is selected, and the average gray value (AGV) of all pixels in this region is calculated.

To classify Baijiu samples of different flavors and ages, PCA, LDA, and HCA methods were utilized to analyze the color information (x, y) and grayscale information (AGV) of the images. Additionally, a 4-layer MLP neural network was used to identify Baijiu adulteration.

For data analysis and software integration, all data were analyzed using Python 3.10.11 programming language with the following libraries NumPy 1.25.2, SciPy 1.11.2, Pillow 10.0.0, Pandas 2.0.3, and Scikit-Learn 1.3.0. For graphical interfaces and data visualization, PyQt 5.15.9, Matplotlib 3.7.2, and Seaborn 0.13.0 were used. Some figures were plotted with OriginPro 2024.

3. Results and discussion

3.1. Identification of baijiu with different flavors

The types and contents of flavor substances determine the flavor of Baijiu, with 12 flavor types of Baijiu available on the market. As shown in Fig. 1A, each Baijiu flavor produces a unique Tyndall light path with differences in brightness and color. Fig. 1B reveals that the AGV of images for sauce and te flavors is higher, whereas the AGV for soybean, rice, feng, and light flavors is lower, leading to darker light paths. Fig. 1C highlights significant color differences in the light paths of various Baijiu flavors sauce and te flavors appear whiter, while laobaigan, feng, and strong flavors lean towards purple. Other flavors exhibit a range of colors between these two extremes.

To validate the above relationship between Baijiu flavor and the grayscale and color of Tyndall effect images, emission spectra for 12 Baijiu flavors were tested, and the test results are shown in Fig. 1D. Sauce and te flavors show much higher spectral values than the other flavors, which is consistent with their brighter light paths. Mixed, sesame, and herbal flavors show moderate spectral values and corresponding brightness, while the remaining seven flavors have lower spectral values and dimmer light paths. Each flavor's spectrum has a distinct main peak position. The spectrum of light flavor Baijiu, for example, features three peaks between 450 and 500 nm, with the highest at 460 nm. For soybean, herbal, sesame, and mixed flavors, the 470 nm peak surpasses the 460 nm peak, becoming the main peak, with 460 nm and 495 nm forming shoulder peaks. In te and sauce flavors, the 460 nm and 495 nm peaks develop into pronounced shoulder peaks, combining with the 470 nm to form a broad peak.

To visually compare color differences, the spectrograms were converted into CIE coordinate diagrams, shown in Fig. 1E. Comparing Fig. 1E and Fig. 1C, the positions of corresponding flavor types are not identical but relatively close. Therefore, this result further proves that the relationship between Tyndall light path color and Baijiu flavor type.

PCA, LDA, and HCA were employed to analyze the color and gray



Fig. 1. Tyndall light path photographs (A), average gray value (B), color coordinate distribution (C), emission spectra (D), emission spectra color coordinate distribution (E) for Baijiu of different flavors.

data. PCA, a dimensionality reduction technique, compresses feature dimensions to analyze key factors' contributions. A larger cumulative percentage with fewer components indicates better performance of the array. In PCA (Fig. 2A), Baijiu samples cluster clearly according to the first two principal components, with distinct separation between clusters. LDA maximizes the distance between categories after projection, distinguishing all Baijiu flavors effectively (Fig. 2B). HCA, an unsupervised classification technique, clusters close samples first, then those further apart, until all samples are clustered into one class (Fig. 2C). These results demonstrate the method's strong discriminative power based on the Tyndall effect, and reveals its great potential for identifying Baijiu flavor types.

3.2. Detection of principal trace flavor compounds

Baijiu contains over 2000 types of trace flavor compounds, which constitute approximately 2% of its total volume. Despite their small percentage, these components play a crucial role in forming Baijiu's 12 distinct flavors. Each compound contributes its unique flavor through mechanisms such as mutual suppression, enhancement, alteration, and blending, together they create the typical characteristics and style of each Baijiu (Dong et al., 2024).

For analyzing flavor compounds, nine trace compounds with concentrations exceeding 200 mg/L in Baijiu were selected. Fig. 3A and Fig. S2 illustrate the optical response of these compounds to a laser beam in the detection setup. Seven compounds (lactic acid, caproic acid, acetaldehyde, acetal, ethyl lactate, ethyl hexanoate, isoamyl alcohol) demonstrate significant optical responses. As shown in Fig. 3B, the light path of caproic acid solution changes with decreasing concentration after laser irradiation, and becomes darker.

The optical response of these pure solutions is primarily due to the fluorescence phenomenon of the substances themselves. Specifically, the molecular interaction of caproic acid and ethanol with water molecules. This interaction forms hydrogen bonds and results in a planar rigid structure, leading to the most pronounced optical response. In contrast, while the concentration of these substances in Baijiu is lower, the light reaction path is brighter due to light scattering phenomena in Baijiu's complex colloidal environment.

To quantitively analyze substance component concentrations, the relationship between the solution concentration of trace components and AGV is modeled as a quadratic function. As shown in Fig. 3C-I, the proposed method, based on the optical response of each solution concentration to the laser, effectively measures the concentration of substance components. The R^2 values of each fitting function exceed 0.9,



Fig. 2. PCA results (A), LDA results (B), HCA Tree Diagram (C) for Baijiu of different flavors.

which confirms the strong correlation between the concentrations of the seven trace components and the AGV of images. These results demonstrate that the proposed method is suitable for identifying the flavor of Baijiu and quantitatively detecting trace compound concentrations.

3.3. Identification of baijiu with different cellar ages

The value of Baijiu increases with the age of cellaring, especially for sauce-flavored Baijiu. To verify the feasibility of the proposed method, sauce-flavored Baijiu of varying cellar age—unaged (0 years), 5, 10, and 15 years—were selected. Fig. 4A displays the images of the Tyndall effect in these Baijiu samples. As observed in Fig. 4B-C, the light path in the 15-year-aged Baijiu is both brighter and wider compared to other samples. In addition, the AGV for the 15-year-aged Baijiu is higher, with its color coordinates closer to white. The emission spectra (Fig. 4D-E) support this observation, and show the highest peak for the 15-year-aged Baijiu and the lowest for the 5-year-aged Baijiu, with the 15-year-aged sample having color coordinates closest to white.

PCA and LDA were used to analyze the color and grayscale data from the Tyndall effect in these Baijiu samples. As depicted in Fig. 5A-B, Baijiu samples of different cellar ages form distinct separate clusters. Furthermore, HCA analysis reveals that samples are preferentially classified by cellar age (Fig. 5C). These findings indicate that Baijiu with the same flavor type but different cellar age exhibits unique characteristics, which are effectively captured by this identification method. The clear separation between these groups demonstrates that the influence of cellar age on the optical response characteristics of Baijiu is significant distinguishable using this method.

3.4. Identification of pure baijiu and its mixtures

Tradition methods struggle to determine whether high-value Baijiu is adulterated with low-value Baijiu (Jia et al., 2023). To address this, we further examined the ability of the presented method to distinguish pure Baijiu from mixed Baijiu. Currently, most high-value Baijiu is sauceflavored, due to the stringent requirements for brewing materials, production methods, and production regions. Moreover, sauce-flavored Baijiu tends to improve in taste with increased cellar age.

In our experiment, a valuable finished sauce-flavored Baijiu, Dongfang Mingzhu Laojiang 15 Baijiu, was selected and mixed in a 1:1 ratio with 11 other flavored Baijiu and 4 sauce-flavored Baijiu. These mixtures were identified using the proposed method.

As illustrated in Fig. 6A, PCA analysis clearly separates pure Dongfang Mingzhu Laojiang 15 Baijiu from each mixed Baijiu, with distinct gaps between clusters. This demonstrates the method's strong identification capability in detecting adulterated Baijiu.

To further test the method's applicability, a 4-layer MLP neural network was employed. The input layer of 3 nodes was used to input the color and brightness information of the Baijiu light path. The output layer of 16 nodes was used to output 16 different results. Two hidden layers, with 4 and 32 nodes respectively, were placed in the middle of the neural network. The test results, represented by the confusion matrix in Fig. 6B, show that all predictions correctly fall on the diagonal. This indicates that there is no misclassification and all Baijiu samples are correctly identified with 100% precision, recall, and F1-score for each sample. This shows the method's robust identification ability for pure and mixed Baijiu samples in practical applications.

3.5. Baijiu analysis and identification system

To facilitate rapid data analysis and enhance user convenience, a Baijiu analysis and identification system was developed using Python and related libraries. Key portions of the codes and comments are provided in the "Supporting Information" section. As showed in Fig. S4, the system is divided into three modules: analysis, construction, and identification. The analysis module is designed to acquire and analyze images. It allows users to capture or import base images, from which the corresponding color and brightness data are calculated and stored. This module also provides a simple visual display of analyzed data. The construction module is responsible for building the relational models. Users can import labeled color and brightness data in batches, which the system uses to generate save the four relational models required for testing. Visual display is also provided for these models. The identification module facilitates the rapid testing of Baijiu samples. Users can capture or import images of the samples to be identified, select the appropriate mode, and load the relevant relational model to obtain the Baijiu identification results.

All data presented in this work were analyzed and processed using this system, which demonstrates excellent practicality and interactivity. In addition, the system's modular design ensures efficient and accurate



Fig. 3. (A) Tyndall effect photograms for 1000 mM of caproic acid (a), lactic acid (b), ethyl lactate (c), acetal (d), acetaldehyde (e), ethyl hexanoate (f), isoamyl alcohol (g), acetic acid (h), ethyl acetate (i), and blank (j). (B) Tyndall effect photograms for 1000, 500, 200, 100, 50, 20, 10, 5, 1, and 0 mM caproic acid. (C—I) Graph of the fitted function of the gray values of the concentrations of caproic acid (C), lactic acid (D), ethyl lactate (E), acetal (F), acetaldehyde (G), ethyl caproate (H), and isoamyl alcohol (I).

Baijiu analysis and identification, which supports both research and practical applications.

4. Conclusions

This work presents a fast and non-invasive method for identifying Baijiu. The method involves illuminated Baijiu with a 405 nm wavelength laser to produce a Tyndall light path. Images of the vertical light path are captured by a camera, and the feature information of these images, such as color coordinates and grayscale values, is analyzed by PCA, LDA, and HCA. Using this method, we successfully identified 12 kinds of flavored Baijiu and 4 kinds of Baijiu with different cellar ages.

We established relationships between the grayscale values and the concentrations of seven trace flavor compounds, which enables the quantitative identification of these concentrations. The method also effectively detects Baijiu adulteration using PCA and a simple identification procedure based on an MLP neural network model. Moreover, the developed analysis platform is user-friendly, and thus facilitates the fast and efficient identification of Baijiu. The method has been proven effective in both experimental and practical applications. It offers an

innovative approach to identify Baijiu reliably as well as a new identification scheme for Baijiu quality control and market supervision.

CRediT authorship contribution statement

Qifei Zhu: Writing – original draft, Software, Methodology, Formal analysis, Conceptualization. Jun Zou: Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. Chunfeng Guo: Writing – review & editing, Project administration, Methodology. Rizeng Tao: Validation, Methodology, Formal analysis. Wenyue Li: Validation, Investigation. Yifan Chen: Validation. Bobo Yang: Resources. Lihua Chen: Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.



Fig. 4. Tyndall light path photographs (A), average gray value (B), color coordinate distribution (C), emission spectra (D), emission spectra color coordinate distribution (E) for Baijiu of different cellar ages.



Fig. 5. PCA results (A), LDA results (B), HCA Tree Diagram (C) for Baijiu of different cellar ages.

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0 0

> F8+DF F9+DF

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0 0 0 0 0

F4+DF F6+DF F7+DF

Predicted labels

0 0 0 0 0 0

F3+DF F5+DF



Fig. 6. PCA results (A) MLP neural network confusion matrix (B) for Baijiu of different samples.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.fochx.2024.101621.

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Q. Zhu et al.

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