



## Multi-Angle Polarization Index System for Pollen Type Bioaerosol Recognition

Qizhi Xu<sup>1,2</sup>, Nan Zeng<sup>1</sup>\*, Wei Guo<sup>1,2</sup>, Jun Guo<sup>1</sup>, Yonghong He<sup>1</sup> and Hui Ma<sup>1,3</sup>\*

<sup>1</sup>Shenzhen Key Laboratory for Minimal Invasive Medical Technologies, Guangdong Research Center of Polarization Imaging and Measurement Engineering Technology, Graduate School at Shenzhen, Tsinghua University, Shenzhen, China, <sup>2</sup>Department of Biomedical Engineering, Tsinghua University, Beijing, China, <sup>3</sup>Center for Precision Medicine and Healthcare, Tsinghua-Berkeley Shenzhen Institute, Shenzhen, China

In this work, we propose a high-throughput online identification method of bioaerosols based on multi-angle polarization index system (MAPIS). In the study, four categories and 10 subclasses of aerosol samples from biological and non-biological sources are detected under three incident polarization mode. Then their measured MAPIS shows that bioaerosols like pollen can be easily distinguished from other types of aerosols. Not only that, experimental results also indicate the feasibility of fine identification between different kinds of bioaerosols based on MAPIS in P and R modes. To further extract simple and optimized polarization characterization parameters suitable for bioaerosols, we analyze the multidimensional data of MAPIS by PCA then validate the aerosol recognition accuracy using the first two principal components by multiple groups of randomly mixed aerosol datasets. The comparison with PCA components based on only scattering intensity demonstrate that MAPIS can be not only applied in the specific identification of bioaerosols but also suitable for the distinction between different kinds of bioaerosols but also suitable for the distinction between different kinds of bioaerosols but also suitable for the distinction between different kinds of bioaerosols but also suitable for the distinction between different kinds of bioaerosols but also suitable for the distinction between different kinds of bioaerosols but also suitable for the distinction between different kinds of bioaerosols.

#### Keywords: polarization scattering, bioaerosol, stokes vector, PCA, pollen

### **1 INTRODUCTION**

Bioaerosols are highly associated with a wide range of health effects with major public health impact [1]. It is important to develop some monitoring system that could offer the capability of real-time monitoring of biological aerosols [2]. Pollen is a major fraction of bioaerosols and is receiving increasing attention due to its high allergenic potential and the associated impacts on personal life quality and economy [3]. Pollens have various effects on human health and the environment. Plant pollens are similarly IgE binding allergens that may cause allergic reactions [4]. Airborne pollens are often considered major agents of allergy-related diseases [5] such as asthma, rhinitis, and atopic eczema [6, 7]. The allergenicity of some pollen is further enhanced by particulate pollution in the atmosphere [8]. Due to the effects of climate change on biota, the negative effects of airborne pollen on humans are increasing [9–11]. The number of people suffering from allergies due to pollen inhalation is increasing every year [12]. Also, for environment, pollen can also act as an environmental pollutant by acting as a nucleus for cloud droplets and ice crystals, affecting the solar radiation reaching earth and the optical properties of clouds, thereby reducing visibility [13].

In the area of public health and allergies, the monitoring and predicting of pollens is challenging, partly due to the lack of standardized and widely applicable offline laboratory analysis or online

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#### \*Correspondence:

Nan Zeng zengnan@sz.tsinghua.edu.cn Hui Ma mahui@tsinghua.edu.cn

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continuous monitoring methods [14]. Traditional pollen monitoring employs fluorescence microscopy, such as extractive staining fluorescence microscopy [15] and direct staining fluorescence microscopy [16]. Moreover, various imaging techniques have been used for pollen detection, such as scanning electron microscopy (SEM) [17], transmission electron microscopy (TEM) [18], x-ray imaging [19], etc. These techniques allow for single particle analysis but provide data at a relatively low time resolution due to time-consuming preparation steps or complicated setups [20–23].

Also, some other methods have been developed based on light scattering, ultraviolet laser-induced fluorescence, and holography combined with deep learning [24]. Wu developed a label-free bioaerosol sensor based on holographic microscopy and deeplearning, which is designed to get rid of transferring to laboratory and manual inspection [25]. Mitsumoto proposed a novel flow particle analyzer based on the design of flow cytometer [26]. The device classifies pollen species by simultaneously detecting both scattered light and the characteristic fluorescence excited by ultraviolet light in the flow cell. Kawashima developed a device which measures the sideward and forward scattering intensities of laser light from each particle to quantify a specific pollen type (Japanese cedar) in Japan [27].

Currently, there are very few related literatures on the polarization characteristics of pollens in ambient air, and the corresponding polarization measurement is only limited to the depolarization rate of pollens [28]. The optical properties of pollen particles can be described by the depolarization rate obtained in the lidar detection [22, 29]. Here non-spherical pollens can produce a strong depolarization rate, which can be distinguished from the background backscattering of other aerosols [22]. In addition, according to the depolarization rate [30–33], many studies have shown the potential to distinguish different kinds of pollens in the atmosphere. There is research work on the Muller matrix of ragweed pollen in the visible spectral range [28], which provides a feasible way to identify pollens by using precise polarimetric fingerprints.

Our previous studies have shown that multi-angle polarization index system (MAPIS) could be used for characterizing nonbiologically derived aerosols such as dust [34], soot [35] or irregular particle samples [36]. In this study, focusing on bioaerosols, we detect the multi-angle polarization scattering signals of individual biological aerosols and then show their characteristic MAPIS different from other abiotic aerosol particles under different incident polarization states. The measured data of various types of aerosol samples are given and compared in this paper, including two dust type samples, two water-soluble type samples, two carbonaceous aerosol samples, and several kinds of bioaerosols (including three kinds of pollens and pearl powder). Each sub-category is measured independently. By principal component analysis (PCA), an unsupervised learning method, we extract some specific indicators based on MAPIS. The results show that, even without the assistance of fluorescence, only by MAPIS, we can accurately distinguish bioaerosols such as pollens from nonbiologically derived particles and can also subdivide the subclasses of pollens. The technology used in this study has

the advantages of non-invasive, online real-time and highthroughput analysis. These preliminary studies confirm the potential of MAPIS applied in a fine identification and characterization of bioaerosols.

#### **2 EXPERIMENTS AND SAMPLES**

#### 2.1 Experimental Setup

Figure 1 shows the schematic diagram of our experimental setup, which has been presented in [37]. The light source is a solid laser (532 nm, 100 mW, MSL-III-532, Changchun New Industries Optoelectronics Technology Co., Ltd.). The incident light can be modulated into three polarization states (horizontal linear, 45° linear and right-handed circular polarization) by PSG (polarization state generator). We define these three measurement modes as H mode, P mode, and R mode. The polarized light is then focused at the center of the air flow by a cylindrical lens. The width and height of the laser spot is 1 and 0.04 mm. In the actual measurement, in order to judge whether any suspended particle is passing through the detection area, we use the intensity at 10° scattering angle as the trigger basis of polarization signal acquisition. When the forward 10° scattering signal exceeds the preset discrimination threshold, the scattered signals at four angles (30°, 60°, 85°, 115°) are then synchronously recorded. For each angle, a spatial filter module composed of a lens and an aperture at fixed location is used to eliminate the influence of stray light. Also, there is an optical trap at the end of laser beam to eliminate the forward stray light.

A four-quadrant polarization state analyzer (0°, 90°, 45°, 135° linear polarizer) is applied at each angle. We also test every fourquadrant polarization module using polarimeter to ensure the orientation deviation of polarizers no more than 2°. The scattered light is spatially divided into four parts and transmitted respectively to four SiPMT detectors via an optical fiber bundle. The light intensity is converted and recorded by data acquisition device (FCFR-USB2068, Fcctec Technology, China) at a sampling rate of 1 MHz. The Stokes parameter elements  $S_{0}$ ,  $S_1$ , and  $S_2$  could be easily calculated as described in **Eq. 1**. Currently we only use linear polarizing films due to the restrictions of the manufacture process, so this study does not involve the circular polarization items. Even so,  $S_1$  and  $S_2$  of Stokes vector at multi-angles already shows ability to characterize bioaerosols such as pollen.

$$S = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} I_0 + I_{90} \\ I_0 - I_{90} \\ I_{45} - I_{135} \\ I_R - I_L \end{bmatrix}$$
(1)

An optimally designed sheath nozzle is used to make sure particles passing through the center of detection area one by one. Sample flow carries sample particles passing through the laser beam within the protection of sheath flow. The effectiveness of the instrument is verified by experiments of standard PSL particles, which has been mentioned in our previous work [38]. The agreement between the measured results and Mie



theoretical calculation results based on a single scattering assumption can further confirm that the multiple scattering is hardly involved in our measurements. The velocity of air flow is controlled by two-flow controller and a gas pump. A particle flies through the detection area within 50  $\mu$ s and we sample one point of signal every 1  $\mu$ s. Thus, our current instrument can obtain signals of up to around 20,000 particles in about 1 s.

#### 2.2 Sample Preparation

We choose four types of typical aerosol samples with different properties: dust, water-soluble salts, carbon, and biologically derived particles. Arizona dust and fly ash are measured as representation of dust. Sodium sulfate and sodium chloride are measured as representation of water-soluble salts. Disordered mesoporous carbon and hollow carbon spheres are measured as representation of carbon aerosols. Chamomile pollen, rose pollen, Osmanthus pollen, and pearl powder are measured as representation of biologically derived aerosols. Each subclass above is measured independently. The Stokes vector elements  $S_0$ ,  $S_1$ , and  $S_2$  are measured at four angles for each measurement mode. Pollen is a common and easily accessible class of biological aerosols. It should be noted that these pollen samples were provided by the drug supplier (Yiqi Herbs), and the pollen went through the grinding process which caused their size to be smaller, but its composition unchanged. According to Ref. [39-42], the untreated pollen size will be larger than 10 um. For example, the diameter of Chamomile Pollen is around 16.6 um [39]. As for pearl powder, it is a mixture of protein ( $\beta$ -chitin, silk-like proteins, and acidic glycoproteins) and calcium carbonate [43, 44], which can also belong to biomass source in composition.

Before measurements, aerosol particles from dust type, carbon type and biological type are screened through a 500-mesh sieve to ensure a relatively uniform particle size and then generated and diffused into uniformly dispersed suspended particles by the TSI-3400A aerosol generator. Salt aerosols of water-soluble type are atomized by a Met One 255 atomizer and then pass through a drying tube. All the detailed morphology information of samples can be found in **Table 1**. The particle size after screening in our experiments is less than 10 microns and was monitored synchronously by an optical particle sizer. In our experiments, we used optical particle sizer (OPS-3330, TSI) for particle size measurement. The measurement process and the accuracy of the OPS can be referenced in [45].

## **3 RESULTS**

# **3.1 Differentiation Between Bioaerosols and Non-Biological Particles**

The Stokes parameters  $S_1$  and  $S_2$  at four angles in each measurement mode for different types of aerosol samples are shown in **Figure 2**. In **Figure 2**, non-biological samples are represented by dots of different colors, while pollen samples are represented by green series cross-symbols. For each subcategory sample, we randomly select 10,000 measured data points to display for convenience. Apparently, compared with the differences within sub-categories of non-biological particles, the difference between non-biological origin samples and bioaerosol samples are significantly larger intuitively in terms of multi-angle polarization index system (MAPIS) regardless of the measurement mode.

Major type	Sub class	Morphology	Diameter	Refractive index
Dust	Arizona Dust	Irregular, diverse shapes from spheres to polygon symmetries [47–49]	1.75 um	1.56–0.026i ((1.56 ~1.65)–i (0.002 ~0.03) [56])
	Fly Ash	Irregular shapes with flaky precipitates or approximately spherical shapes [50, 51]	1.55 um	1.60–0.018i ((1.48 ~1.57)–i (0 ~0.01) [57])
Water Soluble Salts	Sodium Sulfate	Monoclinic, orthorhombic or hexagonal crystal system	0.85 um	1.47-0.002i (1.48-0.001i [58])
	Sodium Chloride	Face-centered cubic	0.57 um	1.50–0.01i (1.54–0.001i [59])
Carbon	Disordered Mesoporous Carbon	Mesoporous material with a disordered structure [52]	0.79 um	1.71–0.212i
	Hollow Carbon Spheres	Hollowed spheres [53, 54]	0.71 um	1.65–0.324i
Biologically Derived	Chamomile Pollen	Prolate-spheroidal, radial symmetry, echinate [39]	1.48 um	1.350–0.012i
Particles	Rose Pollen	Prolate or sub-prolate spheroidal, 3 germ furrows, prominent grooves on the exine surface [40]	1.69 um	1.410–0.020i
	Osmanthus Pollen	Approximately spherical, 3 germ furrows, mesh pattern on the exine surface, slightly wrinkled [41, 55]	1.51 um	1.490–0.022i
	Pearl Powder	Irregular polygonal plate-like structure [42, 44]	2.01 um	1.690–0.046i

TABLE 1 | Morphology of samples.

PCA is defined as an orthogonal linear transformation that transforms the data to a new coordinate system such that the greatest variance by some scalar projection of the data comes to lie on the first coordinate (called the first principal component), the second greatest variance on the second coordinate, and so on [46]. The first principal component can be considered as a projection direction that can best explain the data difference. Then, the ability of the second and third principal components to explain the data difference decreases in turn. Therefore, when we distinguish between biological and non-biological aerosols, the first principal component from the measured data of all kinds of aerosols provides a possible optimal expression for the distinction between these two categories. Similarly, when we further want to accurately identify different subclasses under the category of biological aerosols, the first principal component from the measured data of only various bioaerosols can used as a classification parameter to identify which kind of biological aerosol is detected.

Here we define *X* as a measured multidimensional data matrix, and *w* as a weight coefficient matrix of each principal component. Then the weight coefficient vector of the first principal component,  $w_1$ , can be obtained by optimizing Rayleigh quote.

$$w_1 = \arg \max \left\{ \frac{w^T X^T X w}{w^T w} \right\}$$
(2)

To further extract some specific indicator to distinguish between bioaerosols and abiotic particles, we employ the PCA (Principal Component Analysis) method to analyze the measured MAPIS. The PCA results under different measurement modes are shown in **Figures 3A-C**. PCA is an unsupervised learning method, which means that the input data of PCA does not contain the type information of each particle point. Based on the data distribution along the horizontal axis direction of **Figures 3A-C**, the first principal component extracted by PCA, that is, the direction that shows the overall maximum variance of data, can perfectly separate pollen samples and nonbiological samples. Next, the intra-class differences of pollen samples and non-biological samples are roughly along the vertical axis direction of **Figures 3A–C**, implying that the second principal component probably is suitable for the subdivision of different bioaerosols. The PCA coefficients and interpretation coefficients of the first two principal components for different measurement modes are shown in **Table 2**. Regardless of the measurement mode, the contribution by the extracted first principal component is significantly greater than the contribution of the second principal component.

As a reference, we also use  $S_0$  at four scattering angles in three measurement modes as input for PCA operation. The results are shown in Figures 3D-F, which is similar with the sideward and forward scattering intensities measured in [27]. The PCA results using only multi-angle scattering intensity are quite similar under different incident polarization modes. There is not much difference in the relative positions of different samples, and the difference is likely due to the rotation of the coordinate system. So, the intensity of scattered light from multiple angles alone is not enough to distinguish bioaerosols and non-biological origin particles. However, with the help of MAPIS based on linear polarization vector analysis of the detected light, the high discrimination and specific recognition of bioaerosols can be easily realized. By PCA, we can further extract the first principal components as a good indicator specifically for bioaerosols like pollen.

Concretely, for MAPIS under H mode, we can set the position where the first principal component is equal to -0.5 as the discrimination line, and then determine that the measured data whose value range is on the left of this line comes from biological particles. Similarly, for P mode, the discrimination line can be set at the position of the first principal component equal to 0.4, and for R mode, the line can be set at the position of the first principal component equal to principal component equal to 0.25.

The above discrimination basis can be evaluated on 15 measured datasets which is randomly generated. Each dataset contains measured MAPIS data of bioaerosols and non-biological particles mixed with a certain proportion, and the predicted proportions using the above judgment and the comparison with the preset proportion can be shown in **Figure 4**. We preset five particle number contents of biological aerosols in



the mixed dataset, and then randomly extract data three times for each specified proportion to establish total 15 verification datasets.

For all the 15 datasets, **Figure 4** indicates minor deviations less than 1% between the predicted and true proportion, which further confirms the feasibility of our method to specifically distinguish between bioaerosols and non-biological particles. By an auxiliary observation using a particle size analyzer, there is little difference in the particle size distribution interval of the measured samples. SEM photos of bioaerosols reveal more complex and regular microstructures compared to nonbiological particles. So, the polarization optical difference between non-biological particles and bioaerosols may be due to the microstructures combined with the complex refractive index factor. The relevant detailed microphysical interpretation needs to be further studied.

## 3.2 Fine Subclass Recognition of Bioaerosols

Next, the measured MAPIS of the sub-categories of bioaerosols are shown in **Figure 5**. Compared with **Figure 2**, various Stokes elements at different angles and for different incident polarization states have different recognition abilities. Specifically, the polarization indexes in H mode show a weaker discrimination than those in P mode and R mode. Both the forward ( $30^\circ$  and  $60^\circ$ ) polarized scattering signals in P mode and the backward ( $85^\circ$  and  $115^\circ$ ) polarized scattering signals in R mode seem to be suitable



**FIGURE 3** Pollen and non-biological samples on PCA first and second principal component. (A) MAPIS under H mode; (B) MAPIS under P mode; (C) MAPIS under R mode. (D)  $S_0$  at four angles under H mode; (E)  $S_0$  at four angles under P mode; (F)  $S_0$  at four angles under R mode.

<b>FABLE 2</b>   Principal component coefficients and interpretation coefficien	ts under different mode for pollen and	non-biological samples MAPIS data.
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Mode	PC <sup>a</sup>	30° S1	30° S <sub>2</sub>	60° S1	60° S <sub>2</sub>	85° S₁	85° S₂	115° S₁	115° S₂	IС <sup>ь</sup> (%)
PC2	0.638	0.063	-0.404	0.093	-0.571	-0.230	-0.115	-0.151	1	
P Mode	PC1	0.612	-0.337	0.306	-0.306	0.377	-0.330	0.362	-0.206	81
	PC2	0.306	0.383	0.187	0.347	0.465	0.402	0.209	0.424	15
R Mode	PC1	0.678	0.408	0.136	0.082	0.370	0.267	-0.118	0.353	87
	PC2	0.249	-0.386	0.371	-0.192	0.543	-0.377	0.256	-0.329	9

<sup>a</sup>PC, principal component.

<sup>b</sup>IC, interpretation coefficients.



FIGURE 4 | True and predicted proportion of pollen using PC1 from MAPIS. (A) Measurement conducted in H mode; (B) Measurement conducted in P mode; (C) Measurement conducted in R mode.



TABLE 3 | Principal component coefficients and interpretation coefficients under different mode for biological samples MAPIS data

Mode I	PC <sup>a</sup>	<b>30</b> °	<b>30</b> °	60°	60°	85°	85°	115°	115°	IC <sup>b</sup>	
		S <sub>1</sub>	<b>S</b> <sub>2</sub>	S1	<b>S</b> <sub>2</sub>	S <sub>1</sub>	<b>S</b> <sub>2</sub>	S <sub>1</sub>	<b>S</b> <sub>2</sub>	(%)	
H Mode F	PC1	0.526	-0.409	0.049	-0.177	0.572	0.127	0.034	-0.420	25	
F	PC2	-0.345	0.532	-0.281	-0.307	0.286	-0.154	0.289	-0.484	18	
P Mode F	PC1	0.019	0.185	0.004	0.007	-0.112	0.969	0.009	0.112	64	
F	PC2	0.682	0.103	0.454	-0.090	0.487	0.022	0.265	-0.025	24	
R Mode F	PC1	-0.038	0.237	-0.172	0.385	-0.567	-0.063	-0.228	0.621	60	
F	PC2	-0.134	-0.199	0.662	-0.553	-0.360	-0.019	-0.147	0.210	24	

<sup>a</sup>PC, principal component.

<sup>b</sup>IC, interpretation coefficients.

for the classification and identification of different kinds of biological aerosols.

Similarly, PCA is used to analyze the measured Stokes element  $S_1$  and  $S_2$  at four angles in each incident polarization mode and extract the optimized polarization characterization expression. The PCA coefficients and interpretation coefficients of the first two principal components in different measurement modes are shown in **Table 3**, and the measured data distribution of different biological particles using the first two principal components is shown in **Figures 6A–C**. It can be seen that four bioaerosols are

almost inseparable in H mode but can be clearly distinguished by the principal components in P and R mode. The class separation distances among biological samples using MAPIS in R mode show better discrimination of biological aerosol species than using data in P mode, which implies that particles of biological origin are more sensitive to circularly polarized incident light.

Also, we extract principal components from the scattering intensity  $S_0$  at four angles in three measurement modes for comparison, which is shown in **Figures 6D-F**. Similar with the case in **Figures 3D-F**, the PCA results using only multi-







angle scattering intensity cannot distinguish different kinds of bioaerosols. There is not much difference in the relative positions of the measured data.

Then, we used a verification method similar to that in **Section 3.1**; we constructed five measured datasets of four biological aerosols mixed with different ratios. Based on the statistical distribution on the first principal component for each incident polarization mode, we can predict the proportion of different kinds of bioaerosols and compare them with the preset ratios. The number of aerosol class measured is k;  $x_n$  is the probability density curve of the *n*th aerosol sample on the first principal component. *y* is the measured probability density curve on the first principal component for a mixture of these *k* kinds of aerosols, where *y* and  $x_n$  are vectors. If  $\alpha_n$  is the estimated proportion of the *n*th aerosol class, we multiply and sum the probability density curve for polarization parameters of different kinds of bioaerosols by their proportions, then by fitting the probability density curve based on the estimated mixing ratio of

different aerosol class with the real y based on randomly sampled measured datasets. By least square method in Eq. 1, the optimal solution of proportions could be found out, as shown in Figure 7.

$$\arg \min \left\| y - \sum_{n=1}^{k} (a_n * x_n) \right\|_2 \tag{3}$$

By observing the classification results based on the measured biological aerosol datasets with five different mixture ratios, using MAPIS measured in P and R mode, the identification error of the first principal component is less than 3% for all the datasets. Compared with other abiotic types, the differences of polarization parameters among various bioaerosols are not so big. However, with the help of PCA, different measured Stokes indexes can be combined to form an optimized parameter with a sufficient discrimination suitable for bioaerosol classification.

## **4 CONCLUSION**

In this paper, we investigate the characterization ability of the multi-angle polarization index system (MAPIS) for bioaerosols (especially pollen). Stokes vectors S1 and S2 of 10 kinds of aerosol samples are measured at four scattering angles under three incident polarization states. The types of samples can be divided into four major categories, namely, dust, water-soluble salts, carbon, and bioaerosols. Among them, the first three types belong to non-biological particles, and each of them contains two subclass samples. There are four kinds of bioaerosols, mainly pollens.

Experiment results show that, regardless of the polarization state of the incident light, non-biological particles and bioaerosols can be clearly differentiated based on the measured MAPIS. Moreover, when the incident light is 45° linear polarized or circular polarized, we can also subdivide the kind of bioaerosols according to the data distribution of MAPIS. By comparison with the measured data of multi-angle scattering intensity, the scattering signals without polarization analysis are not sufficient to determine whether the particulate matter is of biological origin or distinguish the sub-categories of bioaerosols. To simplify the multidimensional characterization parameters of MAPIS, the first two principal components extracted by a PCA analysis of all 10 kinds of sample data can be used as specific indicators of bioaerosols. Also, another PCA analysis of four kinds of biological sample data can confirm the feasibility of its first principal component to predict the particle proportion of mixed bioaerosol samples.

To fully obtain and understand the polarization scattering response from more types of biological aerosols, we still have a lot of follow-up work to promote. The limitations of biological aerosol samples in this paper will affect the universality of specific indicators of polarization characterization and related errors. However, the research of this paper still shows the potentials of the synchronous polarization analysis at multi scattering angles. Taking pollen as an example, the microphysical differences between real biological aerosols and abiotic aerosols are difficult to be simply attributed to size or composition factors. Based on the measured MAPIS and the information extraction by machine learning, the accurate

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discrimination and fine classification of biological aerosols like pollen are feasible in on-line high-throughput measurements. The above studies demonstrate the characterization ability of the multi-angle polarization index system (MAPIS) for in situ fast identification of bioaerosols from other non-biological particles. Also, we can subdivide different biological particles based on measured MAPIS of various aerosol samples. PCA analysis can help us extract one or two optimized polarization indexes based on the combination of multiple Stokes vector elements, according to different characterization needs for bioaerosols. Using the first principal component respectively from ten kinds of sample data and four kinds of biological sample data, the specific recognition error of biological type aerosols is no more than 1%, and the discrimination error of different bioaerosols is less than 3%. Our preliminary study lays a solid foundation to further apply polarization technology and method to analyze more important aerosols such as bacteria and virus particles.

## DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

## **AUTHOR CONTRIBUTIONS**

NZ and QX conceived the idea of the manuscript. QX and WG prepare the samples and performed the experiments. QX wrote the original manuscript and analyzed the results. NZ, JG, YH, and HM performed the language editing. All authors have given approval to the final version of the manuscript.

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