



## Scoping studies to establish the capability and utility of a real-time bioaerosol sensor to characterise emissions from environmental sources

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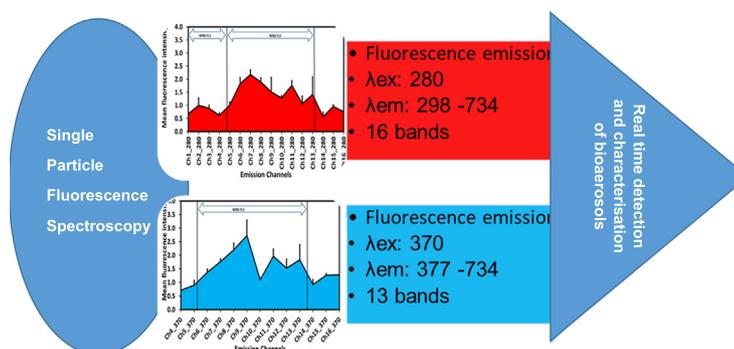
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### HIGHLIGHTS

- First real world evaluation of a novel dual wavelength excitation multiple fluorescence band bioaerosol sensor
- High variability in nature and magnitude of emissions at contrasting sites
- Highly resolved emission intensity measurements provide additional spectral information in comparison to existing devices.
- Differences in emission spectra from different sites at smaller and larger wavelengths than maxima

### GRAPHICAL ABSTRACT



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### ABSTRACT

A novel dual excitation wavelength based bioaerosol sensor with multiple fluorescence bands called Spectral Intensity Bioaerosol Sensor (SIBS) has been assessed across five contrasting outdoor environments. The mean concentrations of total and fluorescent particles across the sites were highly variable being the highest at the agricultural farm ( $2.6 \text{ cm}^{-3}$  and  $0.48 \text{ cm}^{-3}$ , respectively) and the composting site ( $2.32 \text{ cm}^{-3}$  and  $0.46 \text{ cm}^{-3}$ , respectively) and the lowest at the dairy farm ( $1.03 \text{ cm}^{-3}$  and  $0.24 \text{ cm}^{-3}$ , respectively) and the sewage treatment works ( $1.03 \text{ cm}^{-3}$  and  $0.25 \text{ cm}^{-3}$ , respectively). In contrast, the number-weighted fluorescent fraction was lowest at the agricultural site (0.18) in comparison to the other sites indicating high variability in nature and magnitude of emissions from environmental sources. The fluorescence emissions data demonstrated that the spectra at different sites were multimodal with intensity differences largely at wavelengths located in secondary emission peaks for  $\lambda_{ex}$  280 and  $\lambda_{ex}$  370. This finding suggests differences in the molecular composition of emissions at these sites which can help to identify distinct fluorescence signature of different environmental sources. Overall this study demonstrated that SIBS provides additional spectral information compared to existing instruments and capability to resolve spectrally integrated signals from relevant biological fluorophores could improve selectivity and thus enhance discrimination and classification strategies for real-time characterisation of bioaerosols

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from environmental sources. However, detailed lab-based measurements in conjunction with real-world studies and improved numerical methods are required to optimise and validate these highly resolved spectral signatures with respect to the diverse atmospherically relevant biological fluorophores.

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## 1. Introduction

Bioaerosols, airborne particles of biological origin, come from both natural and anthropogenic sources and have potential impacts on global (climatic processes), regional (ambient microbiome) and local (public health) scales. Along with investigations on their climate interaction and long-distance transport, the public health impact of bioaerosols has received significant attention due to a growth in existing and emerging sources of bioaerosols such as waste management operations and intensive agriculture facilities, as well as the resultant human risk of exposure in occupational settings and the wider community (Douglas et al., 2016; Jahne et al., 2016; Madsen et al., 2016; Borlée et al., 2015; Pearson et al., 2015; Walser et al., 2015; Pankhurst et al., 2012; van der Hoek et al., 2012; O'Connor et al., 2010; Bünger et al., 2007; Sykes et al., 2007; Taha et al., 2007, 2006, 2005). Hence there has been increasing interest in detection and characterisation of bioaerosols emissions from various environmental sources. Over the last decade there have been growing efforts to advance methods for detecting the abundance, distribution, diversity and properties of bioaerosols, as well as their environmental impact across different temporal and spatial scales (Anderson et al., 2016; Jahne et al., 2016; Mazar et al., 2016; Pearce et al., 2016; Sialve et al., 2015; O'Connor et al., 2015; Morris et al., 2014; Pankhurst et al., 2012; Vestlund et al., 2014; Sun and Ariya, 2006; Taha et al., 2005).

At present, there are diverse sampling methods with a range of post-collection analyses (culture and non-culture based) for identification and quantification of bioaerosols or their derivatives. However, these are labour intensive and offer snapshot data with low temporal resolution. The capability to quantify the magnitude and spatiotemporal characterisation of bioaerosols emissions from different environmental sources is critical to gauge temporal emission factors and their determinants, developing emission inventories or exposure estimates, advancing forecast modelling and proposing evidence-based management strategies. Recent technological developments have led to the application of a variety of techniques in detection and characterisation of atmospheric bioaerosols from various sources including; electron microscopy epifluorescence microscopy, elastic scattering, laser-breakdown (LIBs), X-ray fluorescence spectroscopy, infrared (IR) absorption, Raman spectroscopy, laser/light-induced fluorescence (LIF), biochemical analysis (e.g., sequencing of DNA or RNA), chromatography, mass spectrometry and nuclear magnetic resonance (NMR) (Pan, 2015; Pöhlker et al., 2012). Among these techniques, fluorescence spectroscopy has shown promising potential for detecting and broadly classifying bioaerosols in real time (Pan, 2015). Instruments based on LIF and/or elastic scattering have recently become commercially available and have shown their capability to detect bioaerosols in real-time over a range of ambient environments and sources: urban/suburban/background (Wei et al., 2016; Yu et al., 2016; Saari et al., 2015; O'Connor et al., 2014; Toprak and Schnaiter, 2013; Gabey et al., 2011; Huffman et al., 2010), dust storms (Hallar et al., 2011), tropical rainforests (Huffman et al., 2012; Gabey et al., 2011; Gabey et al., 2010), high-altitudes (Crawford et al., 2016; Ziemba et al., 2016; Gabey et al., 2013), boreal forest environments (Schumacher et al., 2013; Huffman et al., 2013), industrial processes (O'Connor et al., 2015; Li et al., 2016) and in the atmospheric boundary layer (Perring et al., 2015).

A fairly large body of research is available on lab-based excitation emission characteristics for a range of biologically relevant fluorophores (Hernandez et al., 2016; Pöhlker et al., 2012; Pan et al., 2010; Hill et al.,

2009). However, in the natural environment, bioaerosols are part of a complex mixture differing significantly from lab-based studies. The diversity of biological and non-biological interfering compounds significantly hampers the selectivity of LIF based bioaerosol detectors. The most advanced approach is the use of elastic scattering and dual wavelength excitation of single particles and measurement of spectrally resolved fluorescence along with size and shape in real time. One limitation of existing commercially available LIF based detectors is their broad emission detection bands that make it difficult to classify or discriminate between different types of bioaerosols (Pöhlker et al., 2012). Recently a novel LIF based sensor with highly resolved fluorescence intensity measurements (Spectral Intensity Bioaerosol Sensor (SIBS)) has been developed by Droplet Measurement Technologies Inc. (Longmont, USA). The SIBS is an expansion of the Wideband Integrated Bioaerosol Sensor (WIBS) which was developed by the University of Hertfordshire (Kaye et al., 2005). The WIBS uses two excitation wavelengths ( $\lambda_{ex} = 280$  nm and 370 nm) and measures fluorescence in three emissions ( $\lambda_{em}$ ) bands as follows: FL1:  $\lambda_{ex} = 280$  nm,  $\lambda_{em} \sim 310$ –400 nm, FL2:  $\lambda_{ex} = 280$  nm,  $\lambda_{em} \sim 420$ –650 nm, and FL3:  $\lambda_{ex} = 370$  nm,  $\lambda_{em} \sim 420$ –650 nm. In contrast, fluorescence emission is measured by the SIBS across 16 wavelength bands from  $\lambda_{em} = 288$ –735 nm for two excitation wavelengths ( $\lambda_{ex} = 280$  nm and 370 nm) providing greater spectral resolution in the emission signal from a bioaerosol. In this paper the capability and utility of SIBS was evaluated at contrasting land uses to demonstrate the novel capability of the SIBS to record highly resolved emission spectra. To the best of the authors' knowledge, this is the first study of this kind where SIBS has been employed and tested in a range of real-world emission scenarios.

## 2. Materials and methods

### 2.1. Sampling sites and design

Five contrasting outdoor environments were selected for this study including an agricultural farm, a dairy farm, an urban background site, a sewage treatment works and green waste composting facility (Table 1). All the sites are in the United Kingdom and have been anonymised except for the urban background (Cranfield University). Three measurements were made during daytime at a height of 1 m and site activity logs were kept during each sampling period. Table 1 provides a general description of the sites and sampling strategy.

### 2.2. Instrumentation

Continuous real-time measurements were made with a SIBS comprising of a central optical chamber, a continuous-wave 785 nm diode laser through which particles pass and scatter light, a quadrant photomultiplier tube (PMT) placed to measure the forward scattered light from which the particle shape is derived, an avalanche photodiode (APD) for particle detection and sizing, two pulsed xenon UV sources emitting sequentially at two different wavebands (280 and 370 nm), and a 16 channel photomultiplier spectrometer. A dichroic mirror directs the scattered light to the APD, and the fluorescence emission, 288–720 nm is collected by the two chamber mirrors and delivered through the mirror aperture onto a dichroic beam-splitter that passes the 288–720 nm emissions directly to the spectrometer where it is resolved into 16 channels (Table 2).

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