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Ambient bioaerosol particle dynamics observed during haze and sunny days in Beijing

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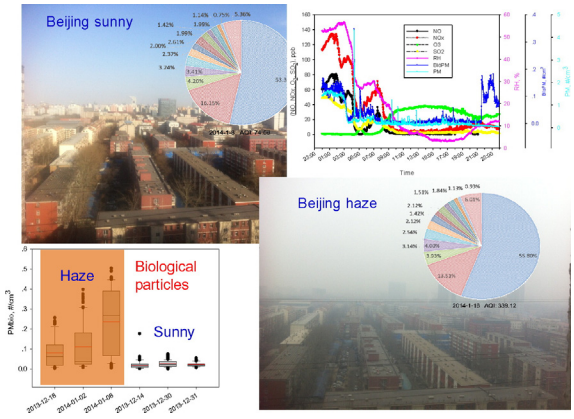
HIGHLIGHTS

- 6-fold higher fluorescent particle concentrations were detected on haze days
- No significant bacterial structure differences between haze and sunny days
- About twice higher endotoxin levels (12.4 EU/m3) were detected on haze days

GRAPHICAL ABSTRACT



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ABSTRACT

The chemical characteristics of airborne particulate matter (PM) have been extensively studied; however, little information exists for its biological components (bioaerosol) especially during a haze event in mega cities. Herein, we studied the bioaerosol (fluorescent particle) dynamics on both haze and sunny days in Beijing from Dec. 2013 to March 2014 by employing a widely used real-time bioaerosol sensor-ultraviolet aerodynamic particle spectrometer (UV-APS). Firstly, we studied the fluorescent particle (BioPM) concentration and size distributions during three independent haze and three independent sunny days. Secondly, we investigated BioPM dynamics over a two-week long monitoring period which included consecutive haze days and alternated sunny days. In addition, we analyzed bacterial community structures and endotoxin levels in the air samples using pyrosequencing and Limulus amebocyte lysate (LAL) method, respectively.

More than 6-fold higher fluorescent particle concentrations up to $5 \times 10^5/m^3$ with peaks at night or early dawn were detected at the time of haze occurrences than those observed on sunny days. When the haze episode progressed for 3–5 days, the BioPM concentrations were observed to decrease to the levels that were typically observed on sunny days. In general, ozone levels were found to be elevated at noon, while BioPM, NO_x and relative humidity were reduced. Gene sequence analysis revealed no significant difference in abundances and

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community structures for top 13 bacterial genera between haze and sunny days, yet about twice higher endotoxin levels (12.4 EU/m^3) were detected on haze days than on sunny days. The results here facilitate a better understanding of atmospheric fluorescent particle dynamics including those under haze events.

1. Introduction

Mankind is facing increased air pollution on a global scale, and extensive studies were carried out in studying the pollution mechanisms and control measures (Rose et al., 2011; Parrish et al., 2011; Seinfeld and Pandis, 2012; Makkonen et al., 2012). However, few studies were conducted to study its biological components, i.e. bioaerosol, including bacteria, fungi, viruses, endotoxin and allergens in heavily polluted mega cities. In a recent study, air samples collected from the automobile air conditioning systems in Beijing were found to contain more than 400 unique bacteria including human pathogens and also pathogenic fungi species, and the bacterial community structures varied with different geographical locations (Li et al., 2013). In addition, it was found that air samples collected in Beijing appeared to have more bacterial diversity than other places such as Hainan and Guangzhou of China (Li et al., 2013). Another study showed that increasing PM concentration resulted in an increased microorganism concentration, and the air samples were found to contain more allergenic and pathogenic materials during polluted days in Beijing (Cao et al., 2014).

Over the past decades, a vast amount of information about ambient bioaerosols has been developed; however the understanding of its atmospheric fate is impacted by the lack of advanced analytical equipment. Equally, this is also due to the fact that only a small fraction of bacterial aerosols, i.e., less than 4%, in the ambient air can be cultured (Tong and Lighthart, 1999; White, 1983). When sampling, their detection can be also influenced by the sampling problems such as impaction and desiccation effects, thus losing culturability and viability (Xu et al., 2013). Recently, there are increased interests in applying the fluorescence based techniques to studying the ambient bioaerosols (Huffman et al., 2010; Agranovski et al., 2003; Hallar et al., 2011; Schumacher et al., 2013; Huffman et al., 2012; Miyakawa et al., 2015; Taketani et al., 2015). One of the instruments applied is Ultraviolet Aerodynamic Particle Sizer (UV-APS). The UV-APS, which was originally designed to detect bio-warfare agents, is a real-time instrument to enumerate fluorescent (often viable bioaerosol in defense community) particle concentration. Its detection of viable bioaerosols is based on the fluorescence emitted by the metabolic products or intermediate products such as pyridine nucleotides (e.g. NAD(P)H) and riboflavin (Harrison and Chance, 1970).

Here, we first chose three independent haze days and three independent sunny days in Beijing to study their respective fluorescent particle (BioPM) concentrations and size distributions by employing the UV-APS. Then, a two-week long continuous monitoring campaign was conducted to further investigate BioPM dynamics during long-term haze days followed by comparison with those of alternated sunny days. Simultaneously, atmospheric NO, NO_x, O_3, SO_2 as well as relative humidity (RH) and temperature were monitored, and air samples were collected and analyzed for their bacterial community structures on haze and sunny days in Beijing using pyrosequencing. Furthermore, the concentration of endotoxin on haze and sunny days were measured using Limulus Amebocyte Lysate (LAL) assay. The results from this work will help understand bioaerosol dynamics during haze days, and also shed light on atmospheric fate of fluorescent biological aerosols.

2. Materials and methods

2.1. Selection of haze and sunny days for monitoring

First, we selected three independent haze days (Dec. 16, 2013; Jan. 2, 2014; Jan. 6, 2014) and three independent sunny days (Dec. 14, 2013; Dec. 30, 2013; Dec. 31, 2013) that occurred in Beijing between December 14th, 2013 and March 5th, 2014 to compare their fluorescent particle concentration levels. Secondly, we conducted a two-week long monitoring campaign from Feb. 21st, 2014 to March 5th, 2014 that included both haze and sunny days (as listed in Table S1) to investigate BioPM dynamics (size distribution, diurnal patterns, species composition) in the atmosphere of Beijing. The sampling site was located on the campus of Peking University (39.9167° N, 116.3833° E) which sits between the intersection of 4th and 5th Rings of Beijing, and the sampling height was about four meters for all measurements. For the first part, we started the monitoring whenever a new haze day occurred. During both campaigns, there was no rain or snow. In this study, the UV-APS (model 3312 A, TSI Inc., MN, USA) was used to generate real-time fluorescent particle size distribution over 24 h for each of the days monitored. For the first part of the investigation, the fluorescent particle concentration levels were recorded every 15 min, and for the second part the data were recorded every minute (for a better time resolution to see the links between chemical and fluorescent components). Through measuring the intrinsic fluorescence (emitted by NADH, NADPH, and riboflavin) level in viable bioaerosol particles, the UV-APS can real-time monitor the size distributions and concentration levels of viable bioaerosol particles in ambient air. For the first part, the sampling started at around 9:00 a.m. every day, and lasted for 24 h for a selected haze or sunny day. For the second part of the investigation, the monitoring continued for the entire sampling period. The air sampling flow rate of the UV-APS at ambient pressure and temperature was 5 L/min including sheath gas flow rate of 4 L/min. Due to instrument or power failures, for some dates or time periods we did not obtain their BioPMs. For second part of the investigation, we have also simultaneously monitored atmospheric NO, NO_x (Chemiluminescence NO–NO_2–NO_x Analyze, Thermo Fisher Scientific, Inc., Waltham, MA), O_3 (UV Photometric O3 Analyzer, Model 49i, Thermo Fisher Scientific, Inc.), SO_2 (Model 43C Trace Level pulsed fluorescence SO2 analyzer, Thermo Fisher Scientific, Inc.) as well as relative humidity (RH) and temperature (Met One Instruments, Inc., Grants Pass, Oregon) for every minute of each monitoring day and studied their possible influences on the PM and BioPM levels.

In parallel, we obtained the air quality index (AQI) values per hour for those selected haze and sunny days and also for the two-week long monitoring from the American Embassy in China (Beijing). The AQI values for 22 typical days are shown in Table S1 (Supporting Information). According to the United States Environment Protection Agency (USEPA), the AQI value is calculated using five criteria air pollutants regulated by the Clean Air Act of the United States, including ground-level ozone, particulate matter (PM), carbon monoxide, sulfur dioxide, and nitrogen dioxide. The range of AQI is 0–500, which is divided into six categories: “good (0–50)”, “moderate (51–100)”, “unhealthy for sensitive groups (101–150)”, “unhealthy (151–200)”, “very unhealthy (201–300)”, and “hazardous (301–500)”. In this study, days with their AQI values below 150 are categorized as sunny days, while haze days are those with AQI values greater than or equal to 150. By this definition, for the second part of the investigation (two-week long monitoring), there were 8 haze days and 4 sunny days in total.

2.2. Bacterial structure and endotoxin studied by high pyrosequencing and LAL assay

In addition to the real-time fluorescent concentration levels, we also collected air samples during haze and sunny days using the button aerosol sampler (SKC, Inc., Eighty Four, PA, USA) in conjunction with mixed