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Diurnal Variations of Size-Resolved Bioaerosols During Autumn and Winter Over a Semi-Arid Megacity in Northwest China

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Abstract Bioaerosols have a major negative effect on air quality and on public health by causing the spread of diseases. This study evaluated the bioaerosol composition and variation in a semi-arid megacity of northwest China from October 2019 to January 2020 using an Andersen six-stage impactor sampler. The size distribution, diurnal variations of the concentrations of airborne bacteria, airborne fungi, and total airborne microbes (TAM) were investigated in autumn and winter. The mean concentrations of airborne bacteria, fungi, and TAM were 523.5 ± 301.1 colony-forming units (CFU)/m^3, 1318.9 ± 447.8 CFU/m^3, and (7.25 ± 1.90) × 10^6 cells/m^3, respectively, in autumn and 581 ± 305.4 CFU/m^3, 1234.4 ± 519.9 CFU/m^3, and (5.96 ± 1.65) × 10^6 cells/m^3, respectively, in winter. The mean bioaerosol concentrations were slightly higher on nonhaze days than on haze days, but the difference was not statistically significant. Higher ambient particulate matter levels and atmospheric oxidation capacity inhibited bacteria survival. The diurnal maximum bioaerosol concentration was observed in the morning in autumn, whereas in winter, bioaerosols did not exhibit such a distribution, the impact of human activities on bioaerosols was still uncertain. The size of airborne bacteria exhibited a bimodal distribution, whereas a unimodal pattern was observed for fungi and TAM. Most bacteria, fungi, and TAM were distributed in the respirable ranges from trachea and primary bronchi to alveoli, indicating that bioaerosols have a high risk of being inhaled and causing respiratory diseases in Xi'an.

Key Points:

- The diurnal pattern of bioaerosol in autumn was clearly different with that in winter
- Airborne bacteria exhibited bimodal distribution while unimodal pattern was observed for fungi and total airborne microbes
- High particulate matter levels and atmospheric oxidation capacity inhibited bacteria survival in winter

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

The concentration of atmospheric particulate matter (PM, e.g., $PM_{2.5}$ and PM_{10}) is a vital index of air quality. In addition to the chemical composition of PM, bioaerosols (airborne microorganisms) have attracted attention from environmental scientists in recent years. Bioaerosols are biological particles in the atmosphere and include bacteria, fungi, pollen, and viruses (Ariya & Amyot, 2004; Jaenicke, 2005; Smets et al., 2016). Bioaerosols can directly influence air quality, cause disease spread (Bush & Portnoy, 2001; Douwes et al., 2003; Ren et al., 2001; Shelton et al., 2002; Xie et al., 2018), and indirectly influence global climate and atmospheric processes (Bowers et al., 2013; Pöschl et al., 2010; Pratt et al., 2009; Qi & Gao, 2006; Sun & Ariya, 2006).

The distribution of bioaerosols varies with seasons, air quality, meteorological conditions, and land use types. Relatively high concentrations of airborne bacteria and total microbes have been detected in Chinese cities, such as Beijing, Qingdao, and Xi'an, in autumn and winter (Dong et al., 2016; Gao et al., 2016; Y. Li et al., 2017), coinciding with the frequent haze events in these cities. However, high concentrations of fungi were found in Beijing in China, Denver and Greeley in the United States, Tijuana in Mexico, Dublin in Ireland, and Seoul in South Korea during summer and autumn (Bowers et al., 2013; Fang et al., 2005; Hurtado et al., 2014; O'Gorman & Fuller, 2008; Uk Lee et al., 2016; Wu et al., 2007). Similar seasonal variations of bacterial and fungal aerosols have been noted in Graz, Austria (Haas et al., 2013), with highest bacterial and fungal concentrations found in winter and autumn. Air quality could be influenced by bioaerosols' distribution. For example, the average concentrations of TAM, viable bacteria, and fungi on haze days ($PM_{2.5}$ ≥ 100 μg/m^3) were 2–5 times higher than those on non-haze days ($PM_{2.5}$ ≤ 100 μg/m^3), because fine particles can act as carriers for microbes and can provide them with nutrition. In addition to the higher fine PM concentration, higher relative humidity and lower solar radiation observed during haze days can also promote microbial growth (Griffin et al., 2003; Y. Li et al., 2015, 2017; Wei et al., 2016; Xie et al., 2018). While

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an inverse relationship between bioaerosol concentration and PM levels has been observed in Beijing, Qingdao, and Xi'an (Gao et al., 2015; Gong et al., 2020; Xie et al., 2018); specifically, bioaerosol concentrations were lower on haze days than on non-haze days. This was probably because of the higher concentrations of heavy metal elements (such as Fe, Pb, and Zn) and polycyclic aromatic hydrocarbons (PAHs) in $PM_{2.5}$ on haze days than on non-haze days (Cao et al., 2018; Hu et al., 2015; L. J. Li et al., 2019), and these toxic chemicals inhibit the growth of airborne microbes. Airborne microbes can attach to PM and undergo long-range transport from their source (Hara & Zhang, 2012; Lee et al., 2009; Raisi et al., 2013). The influence of meteorological condition on bioaerosol distribution were also revealed, which showed high relative humidity leads to an increase in microbes (O'Gorman & Fuller, 2008), whereas high temperatures limit their growth (Y. Li et al., 2017). Núñez et al. (2019) reported that seasonal variations in pollen and fungi are mainly correlated with precipitation.

The aerodynamic diameters of bioaerosol particles govern their distance of spread and infiltration into the human respiratory system, implying variable health risks from microbial inhalation. Much attention has been paid to the size distribution of bioaerosols, as it is closely related to their deposition efficiency and subsidence area and, thus, human health. Bacteria were mainly found to exist in coarse particles (>2.0 μm; Bovallius et al., 1978; Bowers et al., 2013; Cao et al., 2014; Fang et al., 2008; Gong et al., 2020). By contrast, fungi showed a single peak in the fine particle size range, and the particle size of the highest proportion varies among different regions. For example, the distribution of fungi peaked at the ~1.1–3.3 μm PM size in Singapore (Zuraimi et al., 2009), 1.1–4.7 μm in Finland (Hyvärinen et al., 2002), and 2.1–3.3 μm in Beijing and Qingdao (Fang et al., 2008; M. Li et al., 2011). Inhalation is a major entry route for microbial pathogens, and possible adverse reactions can harm both the upper and lower respiratory tract. Respirable bacteria and fungi with a diameter of <4.7 μm can penetrate into the lower respiratory system (Nasir et al., 2012) and elicit allergic or inflammatory responses (Smets et al., 2016). Through endotoxins, bacteria and fungi can produce a strong immune response and can cause acute and chronic health effects (Rylander, 2006).

Xi'an (34.22°N, 109.18°E, 424 m above sea level and 1,100 km from the sea), a typical semi-arid megacity, is located in south margin of the Chinese Loess Plateau. The annual rainfall is ~600 mm. The city has a population of over 10 million and an area of 10,096 km^2. Xi'an has four distinct seasons: Hot summer, cold winter, and comfortable spring and autumn. Air quality in Xi'an becomes the worst in late autumn and winter for winter heating and stable weather (Shen et al., 2008, 2017). The average temperature was 14.7 ± 6.1°C and 2.8 ± 1.6°C in autumn and winter, respectively. Although several studies have evaluated the size distribution of airborne bacteria and airborne fungi over Xi'an (Lu et al., 2019; Qi et al., 2020), limited study have focused on the size distribution and diurnal variations of bacterial, fungi, and TAM simultaneously. In this study, size-resolved bioaerosol samples including airborne bacteria, airborne fungi, and TAM were collected in three periods per day both in autumn and winter (October 2019 to January 2020) in a semi-arid city of Xi'an in Northwestern China. The aims of this study are to (1) determine the diurnal variation of bioaerosols, (2) explore the influence indices of bioaerosols, and (3) examine the size distribution and inhalation risks of bioaerosols. The results of this study could provide a reference for environmental and public health of bioaerosol research.

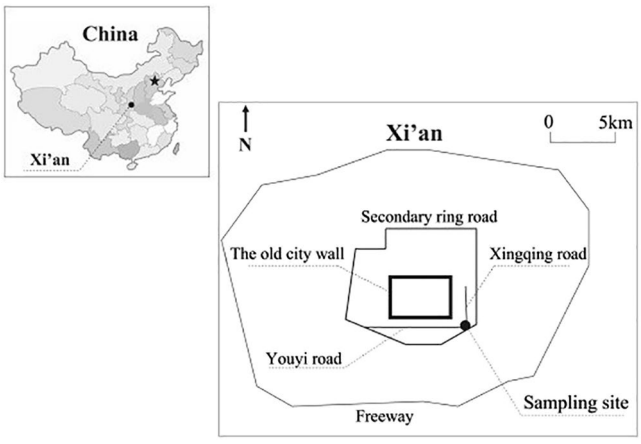

Figure 1. Location of the sampling site.

2. Materials and Methods

2.1. Sampling Site

The sampling site was located in the southeastern part of downtown Xi'an, which is subject to severe heavy air pollution produced by surrounding residential areas and major traffic roads. Sampling was conducted on the roof of a 15 m-high building at Xi'an Jiaotong University (Figure 1). Bioaerosol samples were collected three times per day (8:30, 18:00, and 21:30 of local standard time, LST) from October 30, 2019, to January 14, 2020 (selected 14 sampling days, 7 days in both autumn and

winter). Each sampling time was 10 min both for airborne bacterial and airborne fungi and 30 min for TAM, and 126 samples were collected totally.

2.2. Sampling and Counting Method

Bioaerosol samples were collected using an Andersen six-stage impactor aerosol sampler (Qingdao Juchuang Environmental Protection Group Co., Ltd., Qingdao, China), with a flow rate of 28.3 $Lmin^{-1}$. The sampler was sterilized by 75% ethanol prior to every sampling. Bioaerosols were fractionated into six size ranges and represented different sites (or stages) of the human respiratory system: >7.0 μm (stage 1: Nose and mouth), 4.7–7.0 μm (stage 2: Pharynx), 3.3–4.7 μm (stage 3: Trachea and primary bronchi), 2.1–3.3 μm (stage 4: Secondary bronchi), 1.1–2.1 μm (stage 5: Terminal bronchi), and 0.65–1.1 μm (stage 6: Alveoli; Andersen, 1958). Bioaerosols with aerodynamic diameter <2.1 μm (D_{50} cutoff point of stage 4) and >2.1 μm were defined as fine and coarse particles, respectively.

Culturable microorganism samples (airborne bacteria and fungi) were collected on 9.0 cm Petri dishes containing agar medium for 10 min. Nutrient agar (3 g beef extract, 10 g peptone, 5 g sodium chloride, 15 g agar, 1,000 ml distilled water, pH = 7.2) was used for culturable bacterial samples, and Sabouraud dextrose agar (40 g glucose, 10 g peptone, 20 g agar, 1,000 ml distilled water, pH = 6.2) was used as fungal culture medium (Fang et al., 2008). After sampling, the agar plates were immediately transported to the laboratory for further incubation. Bacterial samples were incubated at 37°C for 48 h, and fungal samples were incubated at 28°C for 72 h. The obtained colony-forming units (CFU) were manually counted after cultivation and were statistically corrected based on the positive-hole correction method (Feller, 1968). The concentration is expressed as CFU/m^3 of air.

TAM samples were collected on sterilized polycarbonate membranes for 30 min (0.2 μm, Whatman, UK). The polycarbonate membranes loaded with bioaerosol were immediately transferred to 250 mL Erlenmeyer flasks containing 30 mL of phosphate buffer (pH = 7.0) and 3 mL of Tween-80 solution, followed by shaking at 120 r/min for 30 min. The samples were then filtered through a black Nuclepore Track-Etch membrane (Whatman) and stained with 10 μg/mL DAPI (4′,6-diamidino-2-phenylindole) for 20 min without sunlight. Next, after samples were stained with DAPI, 10 random fields were selected to count cells using a fluorescent microscope (Olympus, Japan) equipped with an ultraviolet light source (Dong et al., 2016; Xie et al., 2018). The polycarbonate membrane, black Nuclepore Track-Etch membrane, Erlenmeyer flasks, phosphate buffer, etc. all were sterilized at 121°C for 20 min before use. We regularly calibrated the temperature of the autoclave to ensure the sterilization quality by thermometer. The procedures were as follow: Put the thermometer into the autoclave and run the sterilization program. After the sterilization, if the thermometer number is consistent with the setting number of autoclaves, it means that the temperature control of autoclave is good enough to meet the sterilization requirements.

The total airborne microorganism concentration was calculated using the following formula:

$$C = \frac{N_a \cdot S \cdot V_2}{S_f \cdot V_1 \cdot V_3} \quad (1)$$

where C represents the number of total airborne microorganisms in each stage of the sampler ($cells/m^3$), N_a corresponds to the average number of DAPI-stained cells in each field (Cells), S refers to the filtration area on the membrane (mm^2), S_f represents the area of each field (mm^2), and V_1, V_2, and V_3 are the volumes of the filtered sample (L), phosphate buffer (L), and collected air for the sample (m^3), respectively.

Meteorological parameters of temperature (°C) and relative humidity (RH) were measured simultaneously during sampling using a general air quality monitor (JL-03, Qingyi Electronics Co., Ltd., Handan, China). Air quality indices, including PM2.5, PM10, sulfur dioxide (SO2), nitrogen dioxide (NO2), carbon monoxide (CO), and ozone (O_3), were obtained from the Xi'an Environment Protect Bureau (http://www.xaepb.gov.cn/ptl/index.html).

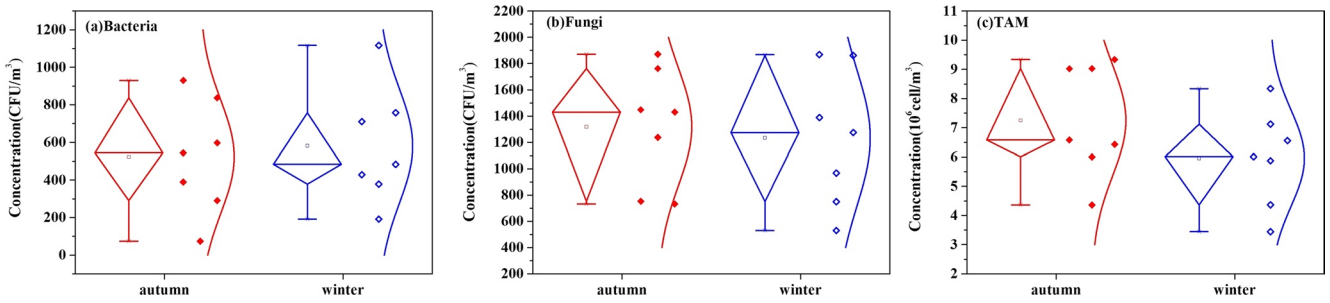

Figure 2. Box plots of seasonal variation of bioaerosols: (a) bacteria, (b) fungi, (c) TAM (total airborne microbe). The lower and upper borders of the box represent 25th and 75th percentile, respectively. Whisker represents the standard deviation. Lower and upper cross marks represent minimum and maximum values, respectively. TAM, total airborne microbes.

2.3. Statistical Analysis

SPSS v22 was used for all statistical analyses. Statistical analysis of variance (AVONA) was used to compare the differences of bioaerosol concentrations between winter and autumn or between haze and non-haze days. In addition, a *t*-test was conducted to compare the diurnal variations of bioaerosol concentrations. A *p*-values less than 0.05 was considered to be statistically significant. Moreover, spearman's correlation analysis was performed to examine the statistical significance of correlation between bioaerosol levels with environmental factors.

3. Results and Discussion

3.1. Bioaerosol Levels Between Autumn and Winter

Figures 2 and S1 illustrate the bioaerosol distribution in autumn and winter. The mean bacterial, fungal, and TAM concentrations were 523.5 ± 301.1 CFU/m^3, 1,318.9 ± 447.8 CFU/m^3, and (7.25 ± 1.90) × 10^6 cells/m^3, respectively, in autumn and 581 ± 305.4 CFU/m^3, 1,234.4 ± 519.9 CFU/m^3, and (5.96 ± 1.65) × 10^6 cell/m^3, respectively, in winter. The mean fungal and TAM concentrations in autumn were 1.1 and 1.2 times higher than those in winter, whereas the mean bacterial concentration in winter was 1.1 times higher than that in autumn. Statistical analysis of AVONA showed that there was no significant difference ($p = 0.607$ [bacteria], $p = 0.792$ [fungi], and $p = 0.087$ [TAM]) of bioaerosol concentrations between autumn and winter. Fungal concentrations were significantly higher than bacterial concentrations (2.5 and 2.1 times higher in autumn and winter, respectively) in both seasons ($p < 0.01$). The proportions of culturable airborne bacteria and fungi in TAM were between 0.7%–1% and 1.8%–2%, respectively. A small fraction of culturable airborne microbes was observed in Xi'an, which is consistent with the findings of M. Li et al. (2011), who reported that the culturable airborne microorganisms accounted for 0.26%–2.04% of TAM in Qingdao, China.

The seasonal variations of both fungal and TAM concentrations in this study were consistent with those in previous studies (Fang et al., 2008; Gao et al., 2016; Haas et al., 2013; M. Li et al., 2011; Y. Li et al., 2017; Qi et al., 2020; Shelton et al., 2002; Yamamoto et al., 2012). For example, a study from Beijing, China, demonstrated that fungal spore concentrations were higher in autumn than in winter, mainly because nutrient levels, plant growth, and weather conditions in autumn favored spore production compared with those in winter (Fang et al., 2008). M. Li et al. (2011) also found that the lowest TAM concentration was in winter in Qingdao, mainly due to low air temperature (−1.3°C). By contrast, studies in Xi'an have shown the highest TAM concentration in winter, which was due to low wind velocity, high PM, and foggy and hazy days (Xie et al., 2018). Moreover, higher airborne bacterial concentration was detected in winter in Beijing, China (Gao et al., 2016), Graz, Austria (Haas et al., 2013), and Punjab, Pakistan (Nasir et al., 2012). The factors influencing bioaerosol composition and concentration are complex because of the variety of microbial species and the combined effect of meteorological and environmental factors in different regions (di Giorgio et al., 1996; Haas et al., 2013; Jones & Harrison, 2004; Uk Lee et al., 2016).

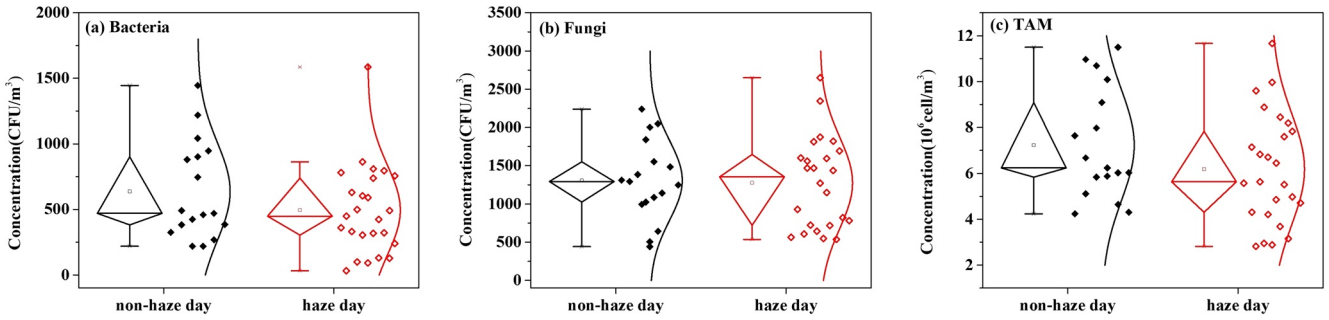

Figure 3. Concentration variations of bioaerosols between haze and non-haze days: (a) bacteria, (b) fungi, (c) TAM. TAM, total airborne microbes.

3.2. Bioaerosol Concentrations Between Haze and Non-haze Days

We explored the variations in bioaerosol concentrations between haze and non-haze days to understand the possible source and potential harm of bioaerosols under different air quality conditions. According to China Meteorological Administration, hazy episodes are generally referred to as days with atmospheric visibility of <5 km, $PM_{2.5}$ value of >75 μg/m^3, and $PM_{2.5}/PM_{10}$ value of ≥60% simultaneously (Technical Regulation for Haze Pollution Day Judging [on trial]). On the basis of these criteria, eight haze days and six non-haze days were identified in the 14 sampling days in this study. The daily mean concentrations of $PM_{2.5}$ were 159.8 ± 67.2 μg/m^3 and 55.9 ± 12.8 μg/m^3 on haze and non-haze days, respectively.

Bioaerosol concentrations between haze and non-haze days were plotted, as illustrated in Figure 3. The mean bacterial, fungal, and TAM concentrations on non-haze days were 30%, 10%, and 20% higher than those on haze days, which are similar to past studies from Beijing, Xi'an, and Qingdao (Gao et al., 2015; Gong et al., 2020). In this study, the difference of bioaerosol levels between haze and non-haze days was insignificant (p = 0.21 [bacteria], p = 0.658 [fungi], and p = 0.175 [TAM]). The air pollutant concentration on haze days increases the toxic and hazardous substances in the ambient air, including crustal elements, heavy metals, inorganic ions, and PAHs (Hua et al., 2015; Kong et al., 2015; Shen et al., 2008; Y. Sun et al., 2006; Z. Sun et al., 2013; Wang et al., 2018), which decrease airborne microbial concentrations. In addition, atmospheric oxidation capacity (O_x), which was calculated as O_3 plus NO_2 concentration, was higher on haze days (95.5 μg/m^3) than on non-haze days (86.4 μg/m^3). A high O_x, as observed on haze days, also inhibits microbial survival (R. Lu et al., 2018).

3.3. Diurnal Variation of Bioaerosols and its Impact Indices

Figure 4 presents the diurnal variation of bioaerosol concentrations in Xi'an. In autumn, the daily maximum airborne bacterial, fungal, and TAM concentrations appeared all around 8:30 (LST), whereas the

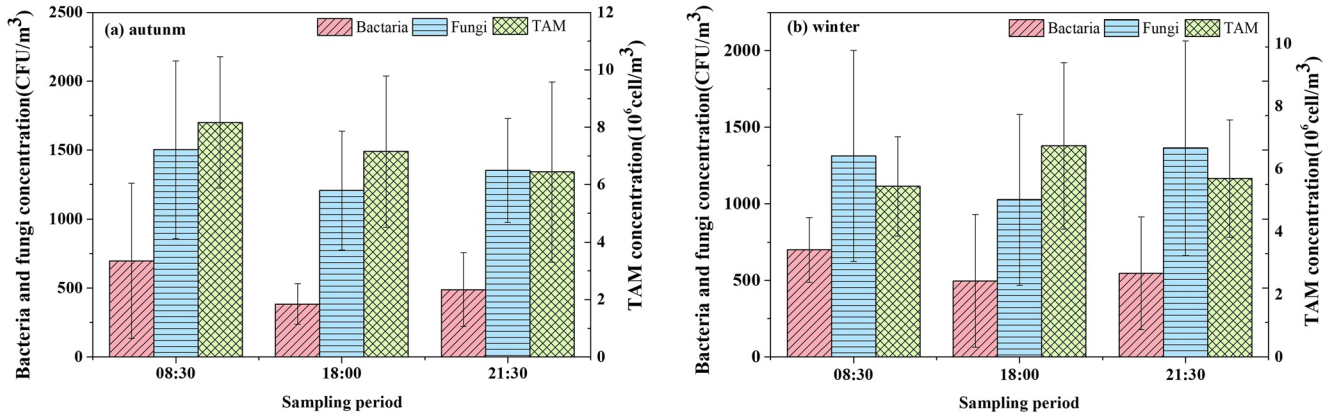

Figure 4. Diurnal variation of bioaerosols in (a) autumn and (b) winter.

daily minimum concentrations of bacteria and fungi appeared around 18:00 (LST), and those of TAM appeared around 21:30 (LST). In winter, the maximum daily concentrations of airborne bacteria, fungi, and TAM in winter were observed at 8:30, 21:30, and 18:00 (LST), respectively, whereas the daily minimum concentrations of TAM were found at 8:30 (LST), and those of airborne bacteria and fungi were observed at 18:00 (LST), which is similar to the autumn variation. There was no statistically significant difference of bioaerosol diurnal levels (In autumn, $p = 0.296$ [bacteria], $p = 0.895$ [fungi], and $p = 0.501$ [TAM]; in winter, $p = 0.537$ [bacteria], $p = 0.589$ [fungi], and $p = 0.484$ [TAM]). Ho et al. (2005) found that the highest total fungal concentration was in the early morning (4:00–6:00 a.m.) and lowest in the afternoon (2:00–4:00 p.m.) in autunm in Hualien, Taiwan. A study reported that the total fungal spore concentration was the highest in the morning in autumn and in the evening in winter (O'Gorman & Fuller, 2008).

Both diurnal distributions of airborne bacterial concentrations and the morning peak in fungal concentrations are attributed to the heat effect of the sun. At sunrise, wind speed and temperature increase, with a decrease in relative humidity, leading to the evaporation of water molecules binding microbial particles to different surfaces. Sunlight in the morning may cause spore release and lead to a maximum peak of fungi in the morning. In the early morning, human activities and traffic flow gradually increase, which disturb and resuspend soil particles into air, potentially increasing airborne microbial concentrations. The decrease in temperature, solar radiation, and traffic rush and increase in human activities are the main causes for the evening peak (21:30 LST) of airborne fungi and TAM. In addition, darkness and high relative humidity in the evening facilitates physical repair of damaged airborne microbes. The diurnal peaks of bioaerosol concentrations can be harmful to human health; further research must analyze the respiratory diseases caused by dominant bioaerosol species.

The diurnal pattern of bioaerosols between haze days and non-haze days exhibited some differences (Figure S2). On non-haze days, bacterial, fungal, and TAM concentrations all peaked at 8:30 (LST), with observed daily minimums at around 21:30 (LST) for fungi and TAM and 18:00 (LST) for bacteria. On haze days, bacterial, fungal, and TAM concentrations peaked at 8:30, 21:30, and 18:00 (LST), respectively. This is consistent with a study in Beijing concluding that fluorescent particle concentrations peak at night or early dawn, when haze occurs, compared with sunny days (Wei et al., 2016). Statistical analysis showed that the diurnal difference of bioaerosol level was insignificant (On haze days, $p = 0.867$ [bacteria], $p = 0.618$ [fungi], and $p = 0.674$ [TAM], on non-haze days, $p = 0.09$ [bacteria], $p = 0.404$ [fungi], and $p = 0.783$ [TAM]) on both haze and non-haze days, which is likely due to the small fluctuation of the diurnal air quality. The influences of air quality and meteorological condition on bioaerosol diurnal distributions were further discussed in the next paragraph.

The impact of environmental indices, including $PM_{2.5}$, O_x, RH, and temperature, on the diurnal variation of airborne bacterial, fungal, and TAM concentrations is plotted in Figure 5. High bacteria concentration was associated with low concentrations of $PM_{2.5}$ and O_x. As PM and O_x concentrations increase, the airborne bacterial concentration decreases due to the pollutant-induced damage to bacteria (Jacumin et al., 1964; Lighthart, 1973; R. Lu et al., 2018; L. Raisi et al., 2010), thereby inhibiting their growth and altering the microbial compositions of PM (Gou et al., 2016; Y. Sun et al., 2018). O_x could damage microbial DNA and cellular structure, thus decreasing the airborne bacterial concentration (C. Fan et al., 2019). However, temperature and RH also play dominant roles in bacteria and TAM distribution (X. Y. Fan et al., 2019; Frankel et al., 2012; Liu et al., 2018; Rajasekar & Balasubramanian, 2011). In addition, spearman's correlation analysis was used to investigate the correlations among various environmental factors ($PM_{2.5}$, atmospheric oxidation capacity [O_x], RH, and temperature) with bioaerosols levels (airborne bacteria, fungi, and TAM). As shown in Table 1, temperature showed significantly negative correlation with bacteria in autumn. Meanwhile, there was a negative correlation between RH and TAM concentration, but temperature had a positive correlation with TAM concentration in winter. The mean ambient temperature was 11.4 ± 3.7°C in autumn and 5 ± 3.3°C in winter. Airborne bacteria had a higher survival rate when temperature was lower in autumn, and relative humidity disfavored the growth of toal airborne microbes in winter. The physiological tolerance hypothesis (Currie et al., 2004), metabolic hypothesis (Allen et al., 2002), biogeography theory, or some combination thereof can explain the temperature–biodiversity relationships; more investigations are warranted. This implies that air pollutants, temperature, and humidity heavily influence bioaerosol formation.

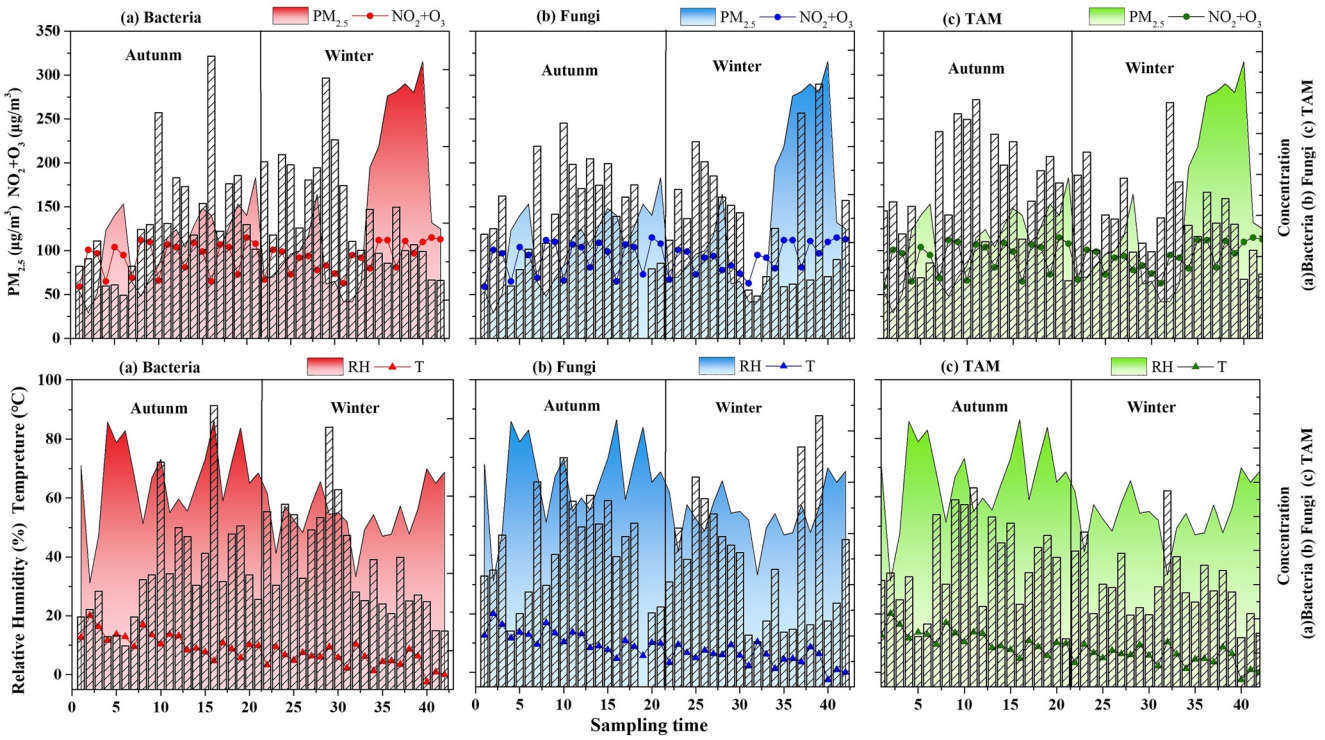

Figure 5. Diurnal variations of $PM_{2.5}$, atmospheric oxidation capacity (NO_2+O_3), RH, temperature (T) with airborne bacteria (a), fungi (b), and TAM (c). PM, particulate matter; RH, relative humidity; TAM, total airborne microbes.

3.4. Size Distribution of Bioaerosols

The size distribution pattern between non-haze and haze days exhibited some differences (Figure 6). The airborne bacteria presented a bimodal size distribution pattern, peaking at >7.0 μm (stage 1) and 3.3–4.7 μm (stage 3). The peak values on non-haze days were much higher than those on haze days, especially in the coarse size peak. It was worth noted that the concentrations of the 0.65–1 μm bacteria fraction declined significantly on haze days, but compared with those on non-haze days, the peak value appeared at 0.65–1 μm from noon to evening (Figure S3), this was mainly due to higher atmospheric acidity, oxidative potential, and reactive oxygen species (ROS) content in PM on haze days (George et al., 2020; Shao et al., 2017; Z. Sun et al., 2014; Zhang et al., 2015), all of which can elicit oxidative stress, cause proinflammatory cytokine release from airway epithelial cells (George et al., 2020), and cause the accumulation of toxic components, thereby promoting particle-induced DNA damage (Shao et al., 2017). Both airborne fungi and TAM exhibited a unimodal distribution pattern, peaking at 1.1–2.1 μm (stage 5). Moreover, the maximum particle size of TAM concentration was 2.1–3.3 μm at morning, and turned to 1.1–2.1 μm at evening on non-haze days. They may attach to the surface of fine PM, leading to an increase in particle size and concentration. Similar to our results, a study in Qingdao indicated that the size distribution of airborne fungi presented a single-peak distribution pattern for all seasons (M. Li et al., 2011). Researchers in Dehradun, India, observed that the total particle concentration for aerodynamic size <1.5 μm was higher than that for particles with larger aerodynamic diameters (Madhwal et al., 2020). Studies have revealed inverse size distribution patterns of bioaerosols that for different environments and regions, that the relevant bioaerosol inhalation risk was different considering the lung deposition convention (Xu & Yao, 2013), and the dominant genera may vary with PM levels (Y. Sun et al., 2018) and seasons (Núñez et al., 2019).

Table 1
Spearman's Correlation Coefficients Between the Concentrations of Bioaerosols and Environmental Factors[a]

	Bioaerosol	$PM_{2.5}$	O_x	RH	T
Autumn	Bacteria	0.139	0.016	0.041	−0.471*
	Fungi	−0.327	−0.09	−0.317	−0.191
	TAM	−0.256	0.012	−0.152	−0.244
Winter	Bacteria	−0.491*	−0.795**	0.121	0.314
	Fungi	0.164	−0.223	0.335	0.155
	TAM	−0.34	−0.165	−0.632**	0.472*

**$p < 0.01$; *$p < 0.05$.
Abbreviations: PM, particulate matter; RH, relative humidity; TAM, total airborne microbes.
[a]The number of samples $n = 21$.

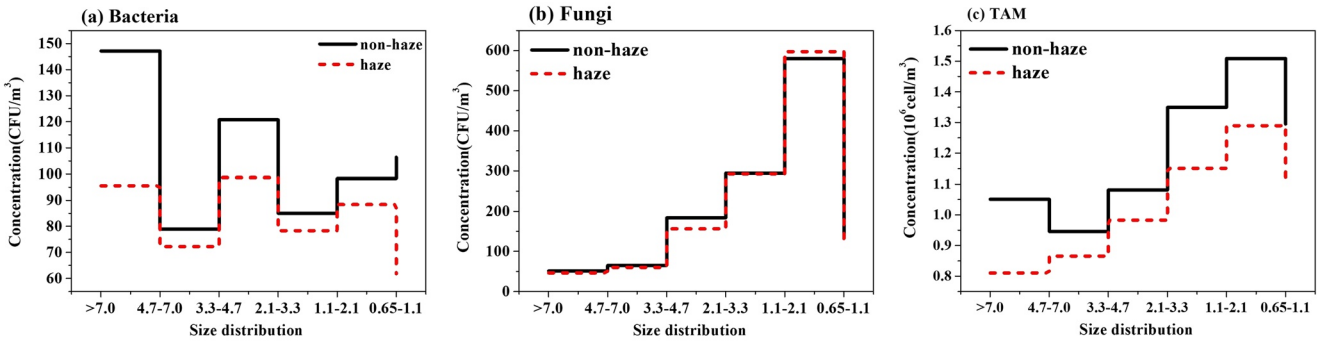

Figure 6. Size distribution of bioaerosols between haze and non-haze days: (a) bacteria, (b) fungi, and (c) TAM. TAM, total airborne microbes.

Compared with the fine PM fraction, airborne bacterial and TAM concentrations were higher in the coarse PM fraction at 67.8% (non-haze days) and 69.7% (haze days) for bacteria and 61.2% (non-haze days) and 61.3% (haze days) for TAM. By contrast, airborne fungi were mainly distributed in fine PM, with a proportion of 54.6% (non-haze days) and 56.5% (haze days) in total airborne fungi. Similar size distributions for bacteria, fungi, and TAM were also found in previous studies (Bauer et al., 2002; Bowers et al., 2013; Cao et al., 2014; Dong et al., 2016; Fang et al., 2008; Gong et al., 2020; M. Li et al., 2011).

Bioaerosols with a diameter of <4.7 μm might easily penetrate the respiratory system. In this study, the inhalable proportions (0.65–4.7 μm) of bacteria, fungi, and TAM were more than 60%, 90%, and 70%, respectively. Notably, the size and composition of bioaerosol particles may be critical in determining their specific biological responses, and the consequent health risk cannot be ignored. Most bioaerosols can be easily inhaled and trigger a series of diseases (Skóra et al., 2015). For example, Kallawicha et al., 2016 found that exposure to fungal spores was positively associated with skin diseases (e.g., atopic dermatitis, contact dermatitis, and other eczematous conditions). Douwes et al. (2003) concluded that fungi and thermophilic bacteria (e.g., Saccharopolyspora rectivirgula and Thermoactinomyces vulgaris; Reboux et al., 2001), which were well-known sources of allergens, may cause hypersensitivity pneumonitis. As the aerodynamic diameter decreases, the primary particle deposition sites in the human respiratory system vary from the trachea and primary bronchi to the alveoli (Andersen, 1958), and the taxonomic composition of the bioaerosols also increases. Future studies should explore the potential relationships between size distribution patterns and microbial compositions of bioaerosol with human respiratory diseases.

4. Conclusions

In this study, the diurnal variation and size distribution of bioaerosols (airborne bacteria, airborne fungi, and TAM) were determined between autumn and winter in Xi'an, China. The bioaerosol concentration in autumn was higher than in winter. Fungal concentrations were much higher than bacteria in both seasons and the proportions of culturable airborne bacteria and fungi in TAM were less than 2%. The mean concentration of bioaerosols on non-haze days was higher than haze days. The diurnal maximum bioaerosol concentration was observed in the morning in autumn. High PM and O_x inhibited airborne bacteria survival in winter. The size distribution patterns of bioaerosols revealed no difference between non-haze and haze days, for airborne bacteria presented a double-peak distribution pattern, whereas the airborne fungi and TAM showed skewed distribution patterns. It was worth especially noting that the proportion of respirable (0.65–4.7 μm) bacteria, fungi, and TAM were much more than 60%, 90%, and 70% of total bioaerosols and presented a high inhalation and respiratory disease risk in Xi'an. The peak value of bioaerosols on non-haze days was higher than that on haze days. Even though the inhalable microbial content in bioaerosols is more than 50%, the proportion of pathogenic microorganisms in that remains unknown. Further studies should focus on the viability and communities of bioaerosols to elucidate health exposure hazards.

Conflict of Interest

The authors declare on behalf of all co-authors that they have no conflicts of interest to this work. The authors declare that they do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

Data Availability Statement

The data of air quality indices were obtained from the Xi'an Environment Protect Bureau (http://www.xaepb.gov.cn/ptl/index.html). The experiment data are publicly available at https://zenodo.org/record/4546078.

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