Evaluation of fungal spore characteristics in Beijing, China, based on molecular tracer measurements

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Abstract

PM₂.₅ (particulate matter with aerodynamic diameters less than 2.5 μm) and PM₁₀ (particulate matter with aerodynamic diameters less than 10 μm) samples were collected by high-volume air samplers simultaneously at a rural site and an urban site in Beijing, China. Various carbohydrates were quantified by high-performance anion exchange chromatography with pulsed amperometric detection (HPAEC-PAD), including the sugar alcohols mannitol and arabitol, recently proposed as molecular tracers for fungal aerosol. The annual average concentrations of arabitol in PM₂.₅ and PM₁₀ at the urban site were 7.1 ± 9.4 and 21.0 ± 20.4 ng m⁻³, and the respective mannitol concentrations were 10.3 ± 9.5 and 31.9 ± 26.9 ng m⁻³. During summer and autumn, higher arabitol and mannitol levels than during spring and winter were observed in coarse particles, probably due to different dominant sources of fungal spores in different seasons. In the dry season (i.e., winter and spring) in Beijing, probably only the suspension from exposed surfaces (e.g., soil resuspension, transported dust, etc) can be regarded as the main sources for fungal aerosols. On the other hand, in summer and autumn, fungal spores in the atmosphere can be derived from more complex sources, including plants, vegetation decomposition and agricultural activity, such as ploughing; these fungal spore sources may contribute more to coarse PM. Moreover, statistical analysis according to typical seasonal patterns, including a dry season (December 2010 to March 2011) and a wet season (July to September 2011), revealed different variations of fungal spores in different seasons. Although fungal spore levels at rural sites were reported to be consistently higher than those at urban sites in other studies, our findings showed the opposite pattern, indicating a high abundance of fungal spores in the urban area of this Chinese megacity.

Keywords: fungal spores, bioaerosol, molecular tracers, mannitol, arabitol, HPAEC

1. Introduction

Biological aerosols are actively and passively released from the biosphere, in which all living things interact, dominating the Earth’s surface and influencing the composition of land, water and air (Després et al 2012). Besides their adverse health effects, biological aerosols play an important role in regulating atmospheric chemistry (Ariya et al 2002). Furthermore, several studies have found that biological aerosols can serve as ice nuclei (IN) and cloud condensation nuclei (CCN), affecting the hydrological cycle and climate change at least on regional scales (Dingle 1966, Schnell...
and Vali 1972, Ariya and Amyot 2004, Sun and Ariya 2006, Christner and Morris 2008, Pöschl et al 2010, Pratt et al 2009). Biological aerosols are transported passively in the atmosphere and many studies have shown that airborne biological aerosols also can be transported on regional or even global and continental scales (Yeo and Kim 2002, Brown and Hovmoller 2002, Prospero et al 2005). Thus, there has been increasing awareness of the biological, chemical and physical effects of biological aerosols in biogeochemistry and atmospheric research, while at present, the components, concentrations and origins of biological aerosols are still poorly understood and quantified.

There are various kinds of biological aerosols in the atmosphere, including fungi, viruses, bacteria, pollen, algae and plant as well as animal debris (Simoneit and Mazurek 1982, Jaenicke et al 2007, Elbert et al 2007). Due to abundant sources (e.g., plants, soil, water, animals and human activities), fungal spores and fragments are found to be one of the most common classes of airborne biological aerosols in many environments (Womololu et al 2003, Elbert et al 2007, Bauer et al 2008b, Crawford et al 2009). Especially in coarse particles, fungal spores were found to be the dominant fraction of biological aerosol components (Glikson et al 1995). The estimated global emissions of fungal spores are ∼50 Tg yr⁻¹, which is comparable with the emissions of anthropogenic primary organic aerosol (∼47 Tg yr⁻¹) (Elbert et al 2007), although higher as well as lower estimates have also been reported (Jaenicke 2005, Winiwarter et al 2009).

The traditional quantitative measurement method for fungi is based on the colony forming units (CFU) assay. This culture method has important limitations, such as in the inability of media to satisfy the specific growth requirements of all fungal species, and only 17% of all known fungal species can be grown by cultivation (Lee et al 2006). Although the dead or uncultivable fungal species in the air cannot form colonies, specific components of them remain toxic or allergenic (Gorny et al 2002, Green et al 2006). Therefore, fungal species—dead or alive, viable or not—can be pathogenic or allergic, and may influence cloud formation and cloud properties. Consequently, the total quantity of fungal aerosols in the atmosphere is usually drastically underestimated by culture-based methods.

To alleviate problems associated with culture-based detection methods, enormous efforts have been made to develop new, more efficient methods for quantifying fungal spores in the atmosphere, including microscopic counting (Bauer et al 2008a), immunoassays (Menetrez et al 2009), molecular tracer methods (Elbert et al 2007), DNA-based approaches (Despré et al 2007, Fröhlich-Nowoisky et al 2009) and real-time detection methods (Schneider et al 2011, Huffman et al 2010). Among these methods, molecular tracers, corresponding uniquely to certain sources or formation processes, can be used to characterize and quantify specific source contributions (Rudich et al 2007). Consequently, molecular tracer methods have been widely used to assess the origins of atmospheric aerosols and are recently also considered as an effective means for evaluating the total load of fungal aerosols, including both viable and dead propagules. For instance, levoglucosan is a common molecular tracer for biomass burning (Simoneit et al 1999), hopanes and steranes for fossil fuel combustion (Rogge et al 1993), odd-numbered (C27–C35) n-alkanes for plant wax (Rogge et al 1991), 3-hydroxy fatty acids for Gram-negative bacteria (Lee et al 2004), etc. Sugar alcohols are a common energy reserve material in fungi, and are produced in large amounts by many fungi. Thus, sugar alcohols, especially arabitol and mannitol, are particularly widespread in fungi and have therefore recently been proposed as molecular tracers for fungal spores (Bauer et al 2008a, Elbert et al 2007) and applied for the assessment of fungal spore contributions to ambient aerosol (e.g., Bauer et al 2008b, Burshtein et al 2011, Claey s et al 2010, Zhang et al 2010, Yang et al 2012). Although other sources of sugar alcohols in airborne particulate matter cannot be excluded, such as plants and algae (Burshtein et al 2011), arabitol and mannitol in the continental aerosols have been mostly associated with fungal spores (Lewis and Smith 1967, Carvalho et al 2003, Ion et al 2005, Jia and Fraser 2011).

The portion of spores in PM$_{10}$ in Amazonia was estimated to be 25% during day time and 45% at night, with an average of 35% (Elbert et al 2007). However, the properties and effects of fungal spores may not only be important in tropical regions, where biological activities are particularly intense (Graham et al 2002, 2003, Gilbert 2005, Elbert et al 2007, Martin et al 2010, Pöschl et al 2010, Zhang et al 2010), but also important for aerosols in urban or suburban areas (Graham et al 2004, Bauer et al 2002, 2008a, 2008b). Bauer et al (2008a, 2008b) applied arabitol and mannitol as tracers for fungal spores and found that fungal spores indeed accounted for a large portion of organic carbon (OC) in the coarse aerosol fraction with a mean value of 60 ± 3% and a considerable portion of coarse PM mass (40 ± 5%) at a suburban site during summer in Mainz, Germany.

China is facing the challenge of serious air pollution, especially in large cities, such as Beijing, which has become one of the atmospheric research hotspots in the world (Chang and Yao 2008), being one of the world’s largest megacities, with a population of over 20 million. Although the annual average PM$_{10}$ levels decreased from 180 µg m⁻³ in 1999 to 132 µg m⁻³ in 2008 in Beijing, the annual average PM$_{10}$ concentrations have been almost constant at ∼130 µg m⁻³ from 2008 to 2011, while being about 30% higher than the Chinese Grade-II standard and seven times the latest standard according to the WHO Air Quality Guidelines (Beijing Municipal Environmental Protection Bureau 2011). High population density results in large sources of fungal spores, since high concentrations of ambient particles (as typically found in densely populated cities) are associated with a high abundance of fungal spores, as was observed in the case of Beijing (Hu et al 2005). Thus, fungal aerosol pollution in Beijing is expected to be more serious than in other cities in the world. In fact, previous studies by traditional culture methods found that the fungal aerosol levels in Beijing were very high (Hu et al 2005). The annual average concentration of airborne fungi in Beijing was observed at 1165 CFU m⁻³ (Hu et al 2005), much higher than those in other places around the world.
(table 1), measured by the same method. With no difference in fungal species, the level of airborne fungal aerosol in the urban area of Beijing was significantly higher than that at an urban location in Nanjing, despite the higher RH and temperatures at the latter location (Zhai et al 2000). Therefore, the objectives of this study were to quantify the fungal aerosol levels in Beijing by the molecular tracer method and investigate the influence of meteorological factors on the concentrations of fungal spores in the urban atmosphere.

2. Experimental procedures

2.1. Sample collection

Samples were collected at two sites in Beijing. One sampling site was on the campus of Tsinghua University (THU), located in the urban area of Beijing. The other site was at Miyun (MY), a rural site, 90 km northeast of the city centre. These two sites have been described elsewhere (Jia et al 2008). Daily PM$_{10}$ and PM$_{2.5}$ samples were collected simultaneously at the THU site from 10 November 2010 to 20 October 2011, except for the days when the instrument required maintenance. The sampling conducted at the MY site included two spring collection periods (8 April to 1 May 2011 for PM$_{2.5}$; 2 May to 12 June 2011 for PM$_{10}$) and two summer periods (13 June to 27 July, 2011 for PM$_{2.5}$; 28 July to 26 August, 2011 for PM$_{10}$). The sampling time of all samples was about 24 h/sample. A total of 329 PM$_{2.5}$ and 283 PM$_{10}$ samples were obtained at THU and 64 PM$_{2.5}$ and 52 PM$_{10}$ samples at MY throughout this campaign. Quality control procedures included collection of field blanks, which were obtained by mounting the filters in the sampler without air flow. Field blanks were collected every ten days during the study period. Both PM$_{10}$ and PM$_{2.5}$ samples were collected by high-volume (Hi-Vol) samplers (GUV-15HBL1, Thermo Fisher Scientific Co., Ltd), equipped with a PM$_{10}$ size-selective inlet or a PM$_{2.5}$ impactor (G1200-41). The nominal flow rate of the Hi-Vol samplers was held constant at 1.13 m$^3$ min$^{-1}$. All PM$_{10}$ and PM$_{2.5}$ samples were collected on quartz fibre filters, prebaked at 550°C for at least 8 h to remove organic material. The filters were stored at −20°C after sample collection.

2.2. Chemical analysis

A punch (2.2 cm$^2$) of each quartz filter was extracted with 2.0 mL of ultra-pure deionized water (＞18.2 MΩ resistivity) under ultrasonic agitation for 60 min. The aqueous extract solutions were filtrated through syringe filters (0.45 μm, Pall Corporation, NY, USA) to remove insoluble materials. All sample extract solutions were stored at 4°C until sample analysis. Arabinol, mannitol and other carbohydrates were quantified by high-performance anion exchange chromatography with pulsed amperometric detection (HPAEC-PAD) on a Dionex ICS-3000 system (Dionex, Sunnyvale, CA, USA), which consisted of a dual pump module, detector/chromatography compartment and autosampler. The separation was carried out on a Dionex CarboPac MA1 analytical column and guard column with an aqueous sodium hydroxide (NaOH, 480 mM) eluent at a flow rate of 0.4 mL min$^{-1}$. All carbohydrate concentrations were below detection limits in the field blanks. More details about the HPAEC-PAD method can be found elsewhere (Linuma et al 2009).

2.3. Meteorological data

The meteorological data were obtained from the China Meteorological Data Sharing Service System (http://cdc.cma.gov.cn/), including temperature, relative humidity (RH), sunshine duration and wind speed. During the campaign, the annual average temperature, RH, wind speed and sunshine duration were 13.5 ± 11.6°C, 46.3 ± 20.7%, 2.3 ± 1.0 m s$^{-1}$ and 8.0 ± 2.9 h, respectively.

3. Results and discussion

The annual average concentrations of arabinol in PM$_{2.5}$ and PM$_{10}$ at the THU site were 7.4 ± 9.4 and 21.0 ± 20.4 ng m$^{-3}$, while the respective mannitol concentrations were 10.3 ± 9.5 and 31.9 ± 26.9 ng m$^{-3}$. Figure 1 shows the daily concentrations of arabinol and mannitol in PM$_{10}$ and PM$_{2.5}$ from 10 November 2010 to 20 October 2011 measured at the urban site (THU). In PM$_{10}$, the maximum values of airborne arabinol and mannitol were 133.0 and 137.8 ng m$^{-3}$, and those in PM$_{2.5}$ were 74.2 ng m$^{-3}$ and 95.3 ng m$^{-3}$, respectively. The variations in both arabinol and mannitol concentrations in PM$_{10}$ and PM$_{2.5}$ were fairly consistent, as seen in the time series plots (figure 1). Compared to PM$_{10}$, the monthly average concentrations of arabinol and mannitol in PM$_{2.5}$ did not vary significantly and were present at nearly
consistent levels in the different seasons (figure 2). During the summer (June to August) and autumn (September to November) higher arabitol and mannitol levels were observed than during spring (March to May) and winter (December to February) (figure 2). This pattern was more pronounced in coarse particles (as reflected in the PM$_{10}$ values), i.e., in summer and autumn, the fungal spore tracers were much more abundant in the coarse mode, while in spring and winter the tracers were distributed more evenly between coarse and fine particles. Similar observations have also been reported by Yttri et al (2007), who found that sugar alcohols, including arabitol and mannitol, were more abundant in fine aerosols during winter, with a smaller contribution in summer. One possible reason for this pattern is different dominating sources of fungal spores during the different seasons. In winter and spring, i.e., the dry season in Beijing with fewer plants and no agricultural activity, the suspension from exposed surfaces is the main source for fungal aerosols, resulting in higher PM$_{10}$ fractions of arabitol and mannitol (figure 2). On the other hand, in summer and autumn, fungal spores in the atmosphere can be derived from much more complex sources, e.g., plants, vegetation decomposition and agricultural activities, such as ploughing, which may contribute more to coarse PM. High fungal spore levels during summer and autumn were also found in other studies (Kourtchev et al 2011, Burshtein et al 2011, Yttri et al 2007, Lau et al 2006).

The correlations between the concentrations of the two sugar alcohols in PM$_{2.5}$ and PM$_{10}$ at THU are shown in figure 3. In general, mannitol and arabitol correlated well with each other in both PM$_{10}$ ($R^2 = 0.71$) and PM$_{2.5}$ ($R^2 = 0.81$) (figures 3(a) and (b)). However, the relationship between mannitol in PM$_{2.5}$ and PM$_{10}$ was relatively weaker ($R^2 = 0.52$) (figure 3(d)), which was probably due to different dominant sources of fungal spores in different PM size fractions. In the fine PM fraction the sources of fungal spores appear to have been slightly more similar, while in coarse particles, the fungal spores may have been derived from more complex sources, such as plants, vegetation decomposition and agricultural activities, as discussed above. This observation also agrees with our findings of higher
concentrations of fungal spores in coarse PM during summer and autumn compared to the other seasons. To better evaluate the seasonal patterns in the sources of fungal spores, statistical analysis of mannitol in PM$_{2.5}$ and PM$_{10}$, was further based on typical dry season (December 2010 to March 2011) and wet season (July 2011 to September 2011) periods (figure 4). In the dry season, the concentrations of mannitol in PM$_{2.5}$ and PM$_{10}$ correlated well with each other ($R^2 = 0.77$), while there was no correlation in the wet season ($R^2 = 0.31$) (figures 4(a) and (b)). Similarly, the correlation between arabitol in PM$_{2.5}$ and PM$_{10}$ in the dry season ($R^2 = 0.87$) was also significantly higher than that in the wet season ($R^2 = 0.34$) (data not shown). The high correlation between mannitol in PM$_{2.5}$ and PM$_{10}$ indicates a simpler source spectrum of fungal spores in the dry season, while the poor relationship in the wet season reveals more complex sources for fungal spores under those conditions. However, different patterns of arabitol and mannitol concentrations in PM$_{2.5}$ and PM$_{10}$ were found at Jianfengling Mountain, a tropical rainforest in China (Zhang et al 2010), probably due to the different type of location (tropical rainforest) and the associated fungal habitats, compared to our study, which was conducted in an urban area. The two sugar alcohols in the coarse PM fraction in the tropical rain forest were mainly derived from the same sources, i.e., plants, while in fine aerosol, the poor correlation was likely due to the influence of additional biogenic sources of fungal spores.

The temporal variations of arabitol concentrations at THU and MY were fairly consistent in the different seasons, as shown figure 5. According to other studies, fungal aerosol
levels at rural sites are typically higher than those at urban locations (Lau et al. 2006, Yttri et al. 2007, Bauer et al. 2008a, 2008b). Consequently, the arabitol levels at MY, being a rural site with land cover dominated by shrubbery, were expected to be significantly higher than those at the urban site (THU). However, during most of the study period, the ambient levels of arabitol in PM$_{2.5}$ and PM$_{10}$ at THU were not lower than those at MY (figure 5), revealing an unexpectedly high abundance of fungal spores in the urban area of Beijing. For example, during the summer season, the arabitol concentrations in PM$_{2.5}$ at MY were lower than those at THU, while the ones in PM$_{10}$ were higher than those at THU at times (figures 5(c) and (d)). This pattern was likely due to the stronger fungal activities contributing to coarse PM in that period, associated with larger vegetation coverage during the summer time in the MY area. The different pattern of arabitol concentrations at MY and THU from 16 July 2011 to 1 August 2011, was possibly caused by the influence of long-range transport of fungal spores from distant sources during that period. Pollutants, along with airborne biological material, were transported from south and southwest China, e.g., Hebei province, with more humid and warm conditions, by the prevailing southerly winds in that period, as supported by air mass back trajectory analysis (data not shown). The MY site was situated to the northeast of Beijing and surrounded by highlands on three sides, except the south, resulting in the accumulation of arabitol (and other aerosol species) advected from the source areas to the south.

Figure 6 shows the correlations between arabitol concentrations in PM$_{10}$ and various meteorological parameters at the THU site. The meteorological conditions can significantly affect the initial release and dispersal of fungal spores in the atmospheric environment. Temperature and water availability can influence the magnitude of the source and control the release of fungal spores, while sufficiently strong air movement or mechanical disturbance is also needed for fungal spores to be released from the surface (Jones and Harrison 2004). In our study, the arabitol concentrations in PM$_{10}$ exhibited positive correlation with average temperature, which was reported for several previous studies as well (Zhang et al. 2010, Sousa et al. 2008, Ho et al. 2005). Higher levels of arabitol were present at times, coinciding with higher daily temperatures, especially on days with temperatures ranging from 20 to 30$^\circ$C, whereas in case of daily temperatures below 0$^\circ$C, the arabitol concentrations were the lowest (figure 6(a)). Lin and Li (2000) observed the maximum concentrations of fungi when ambient temperatures were 25–30.8$^\circ$C and relative humidity was 60–70%. The ambient concentrations of arabitol obtained from this study increased with RH below 70%. The ambient concentrations of arabitol obtained from this study increased with RH below 70%. High arabitol abundance (30.0 ng m$^{-3}$ on average) was associated with an RH range of 51–70%, while the lowest arabitol concentrations were observed under dry conditions (RH < 30%) with an average of 7.3 ng m$^{-3}$ (figure 6(b)). There was an obvious decrease in arabitol levels at RH higher than 70%, which was consistent with previous observations as well (Lau et al. 2006), confirming that the release of fungal spores from their substratum can be depressed by more humid conditions (RH > 70%).
Figure 6. Correlations between arabitol concentrations and (a) temperature, (b) relative humidity, (c) wind speed and (d) sunshine duration in PM$_{10}$ at THU.

Wind speed is an additional important factor influencing the release of fungal spores into air. Fungal spores require sufficient wind speed to be removed from a surface, while the dispersion of fungal spores also increases with higher wind speeds. Lin and Li (2000) found that the ambient fungal spore concentrations reached peak values when wind speeds were less than 1 m s$^{-1}$ in Taipei. In this study, the wind speed was in the range of 0.6–6.3 m s$^{-1}$, and the highest ambient fungal spore concentrations were also observed at a wind speed range of 0.6–1.0 m s$^{-1}$ with an average of 25.0 ng m$^{-3}$ (figure 6(c)). In general, the arabitol concentrations in Beijing exhibited negative correlation with wind speed. A correlation analysis of the wind speed and concentrations of fungal spore tracers was done for the rural site as well. Although the surrounding environment at the rural site was different from the urban site, the arabitol concentrations also exhibited negative correlation with wind speed at MY (data not shown).

Solar radiation can influence the levels of ambient fungal spores as well. Su et al (2000) reported that the release of fungal spores can be affected by the duration of light exposure. However, solar radiation is measured only at a limited number of sites in the world, while sunshine duration is measured in many stations (Lam and Li 1996). Sunshine duration is defined to be the sum of all time periods during the day when the direct solar irradiance equals or exceeds 120 W m$^{-2}$ (Yorukoglu and Celik 2006). Global solar radiation can be well correlated with sunshine duration and many regression techniques have been used to investigate the correlations of solar radiation with sunshine duration on regional and global scales (Lam and Li 1996, Yorukoglu and Celik 2006, Li et al 2011). Consequently, sunshine duration has been the most widely available measure for solar radiation estimations. Therefore, in this study, daily sunshine duration was used as an indirect indicator of solar radiation intensity. Unlike the correlation between temperature or wind speed, ambient concentrations of the fungal spore tracers exhibited no evident relationship with sunshine duration during the four seasons (figure 6(d)), possibly due to different influence on fungi with the same sunshine duration in the dry and the wet seasons. Thus, the statistical analysis of arabitol in relation to sunshine duration was further based on typical dry season (December 2010 to March 2011) and wet season (July 2011 to September 2011) data, revealing different patterns in the two seasons (figure 7). Arabitol concentrations in the wet season exhibited no relationship with sunshine duration (figure 7(b)), while in the dry season an evident negative relationship was observed (figure 7(a)). This phenomenon may be due to the fungi activity being more strongly affected by sunshine duration under low ambient humidity (i.e., in the dry season), while in humid environments, the tolerance of fungi to sunshine exposure may be increased, resulting in the absence of any relationship between fungal activity and sunshine duration in the wet season.

4. Conclusions

Fungal spores, the most abundant class of biological aerosol in the atmosphere, have significant adverse health effects and
also play important roles in regulating certain atmospheric processes. The polyols mannitol and arabitol, recently proposed as tracers for fungal spores, were measured in \( \text{PM}_{2.5} \) and \( \text{PM}_{10} \) in Beijing, China. The annual average concentrations of arabitol in \( \text{PM}_{2.5} \) and \( \text{PM}_{10} \) at an urban site were \( 7.4 \pm 9.4 \text{ ng m}^{-3} \) and \( 21.0 \pm 20.4 \text{ ng m}^{-3} \), while the respective mannitol concentrations were \( 10.3 \pm 9.5 \text{ ng m}^{-3} \) and \( 31.9 \pm 26.9 \text{ ng m}^{-3} \). During summer and autumn, arabitol and mannitol levels were higher than in spring and winter, mostly pronounced in coarse particles. This was probably due to different dominant sources of fungal spores in different seasons. In the dry season (i.e., winter and spring) in Beijing, the suspension from exposed surfaces can be regarded as the main source for fungal aerosols, while in summer and autumn fungal spores in the atmosphere can be derived from more complex sources, such as plants, vegetation decomposition and agricultural activity, which contribute more to coarse PM. Further seasonal statistical analysis revealed different variations of fungal spores in the dry season and wet season. The arabitol levels at the urban site were higher than those observed at the rural site, in contrast to previous observations, indicating that fungal spores were highly abundant in the urban area of Beijing. In addition, meteorological conditions (e.g., temperature, relative humidity, wind speed and sunshine duration) were shown to have complex effects on the concentrations of fungal spores in the atmosphere. As airborne fungal spores play an important role in the formation of clouds and ice nuclei and affect the spread and reproduction of organisms in the biosphere, these effects need to be further investigated even (or especially) in urban areas, as demonstrated in this study for Beijing, China.

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