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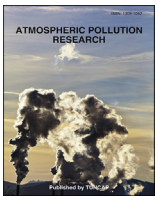

Original Article

Contribution of fungal spores to organic carbon in ambient aerosols in Beijing, China

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ABSTRACT

Fungal spores are ubiquitous components of atmospheric aerosols and contributors to the organic carbon (OC) component in ambient aerosols. In order to better understand the role of fungal spores and their impact on atmospheric processes, this study was conducted to investigate the contribution of fungal spores to OC at urban and rural sites in Beijing, China. Ambient concentrations of a molecular tracer for fungal spores, i.e., mannitol in PM_{10} and $PM_{2.5}$ samples were measured at an urban site (Tsinghua University, THU) during an entire year, while the observations in PM_{10} at a rural site (Miyun, MY) were conducted during late spring and summer. Combined with the factor representing the average content of mannitol per spore (0.49 ± 0.20 pg) obtained at the same urban site in Beijing, the year-round number concentrations of fungal spores were obtained. Using a conversion factor of 13 pg C spore^{-1}, the annual average concentrations of spore-OC in $PM_{2.5}$ and PM_{10} at the THU site were observed at 0.3 ± 0.2 μgC m^{-3} and 0.8 ± 0.7 μgC m^{-3}, while the respective contributions of spore-OC to total OC were 1.2 ± 0.7% and 3.5 ± 3.7%, respectively. The contributions of fungal spores to OC in the two size fractions had the following seasonal trend (from highest to the lowest levels): summer, autumn, winter and spring. During the summer sampling season, the contribution of fungal spores to OC was observed at a higher level at the rural site (14.1 ± 10.5%), compared to the urban site (7.3 ± 3.3%). It can be concluded that fungi are a non-negligible source of carbonaceous aerosol even at urban locations such as Beijing, China. Thus, more studies are needed to better understand the spatial, temporal and size distributions of fungal OC contributions to atmospheric aerosols in populated areas.

1. Introduction

Organic aerosol (OA), an aggregate of thousands of individual compounds, constitutes a significant fraction of atmospheric aerosols. Depending on geographical locations, OA accounts for 10–80% of the total $PM_{2.5}$ mass (Turpin and Lim, 2001; Ho et al., 2014; Wang et al., 2015a), and as high as 90% in the Amazon region (Kanakidou et al., 2005). With the rapid pace of urbanization and industrialization, Beijing has become one of the atmospheric research hotspots, and numerous studies have been carried out in the past decade on chemical characteristics of PM, especially for organic components (Chan and Yao, 2008; Liang et al., 2012; Cheng et al., 2014; Tan et al., 2016). Studies have revealed OA to account for 10–25% of the total $PM_{2.5}$ mass in Beijing (Duan et al., 2005; Wang et al., 2015a, 2015b; Guo, 2015). Wang et al. (2015a) found that carbonaceous materials accounted for 17.3–21.2% of the $PM_{2.5}$ in the Beijing-Tianjin-Hebei Region, with a much higher content under haze conditions. Furthermore, Xu et al. (2015) reported that organic compounds were observed

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to be even higher with 39.9% of PM mass in the <0.43 μm size range at an urban site in Beijing.

Detailed speciation of the organic compounds and their size distributions can provide important information on roles of organic carbon (OC) in climate modulation, health implications, visibility reduction and chemical reactions in the atmosphere. However, substantial amounts of atmospheric OC remain uncharacterized and unidentified (Cheng et al., 2009). The sources of OC emissions can be divided into anthropogenic and biogenic. Most of the air pollution studies in Beijing have focused on organic components of aerosols associated with anthropogenic sources (e.g., Li et al., 2013; Tan et al., 2016), while little research was focused on biogenic emissions among the massive aerosol burden in Beijing.

Primary biological aerosol particles (PBAPs) are ubiquitous in the atmosphere, comprised of diverse types of biological substances such as fungal spores, bacteria, viruses, pollen, skin fragments, plant debris, etc (Després et al., 2012). Microbiological substances are conventionally considered as a minor OC source of aerosols. However, recent studies found that PBAPs can contribute a significant fraction to total aerosol particles in terms of number concentrations. Graham et al. (2003) observed that particles smaller and larger than 1 μm comprised up to 40% and 80% of the total aerosol number concentration of PBAPs measured in a tropical forest, respectively. Biological particles accounted for up to ~50% of total particles at a semi-rural location in Mainz, Germany, and ~20% at a remote continental location at Lake Baikal, Russia, as reported by Jaenicke (2005).

Due to biogenic 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 species in different regions (Bauer et al., 2008a, 2008b; Liang et al., 2013a; Zhang et al., 2010, 2015). 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). Therefore, fungal spores can be a significant source of OC in ambient aerosols. Elbert et al. (2007) estimated that fungal spores accounted for an average of 35% of the $PM_{1\text{-}10}$ mass in Amazonia. Employing the conversion factor of 13 pg carbon per fungal spore, Bauer et al. (2008b) estimated about 14% of OC in PM_{10} to be from fungal spores at a suburban site in Vienna. Womiloju et al. (2003) estimated that fragments of fungi and pollens collectively accounted for 12–22% of the total OC in the fine particulates ($PM_{2.5}$) in Toronto, Canada, using phospholipid tracers. Global model simulations also suggested that fungal spores can account for up to 23% of total primary emissions of organic aerosols with the highest abundance in tropical rainforests (Heald and Spracklen, 2009). Furthermore, fungal spores were found to be the dominant fraction of biological aerosol components in coarse particles (Glikson et al., 1995; Taylor et al., 2002; Zhang et al., 2010, 2015). Based on the tracer method and principal component analysis, Zhang et al. (2015) reported ~66% of OC in coarse particles was contributed by fungal spores in the Jianfeng mountain (JFM) natural reserve on Hainan Island, China.

Understanding fungal spore contributions to OC and their size distributions will certainly facilitate better comprehension of their role and impact on atmospheric processes. Our former research has revealed that fungal spores in Beijing were present at concentration levels which were noticeably higher than those in urban areas in Europe and the US (Liang et al., 2013a). Nevertheless, there is little known about the contribution of fungal spores to OC in this region. In light of these research gaps, this study was conducted to investigate the contribution of fungal spores to OC, as well as to describe seasonal variations and comparison of urban and rural sites in Beijing, China.

2. Material and methods

2.1. Aerosol sampling

Samples were collected at two sites in Beijing. One sampling site was on the campus of Tsinghua University (THU), where two samplers were placed on the roof of a 3 m tall building, surrounded by teaching buildings and dormitories. Daily PM_{10} and $PM_{2.5}$ samples were collected simultaneously at the THU site from 2011/11/10 to 2011/10/20. The other site was at Miyun (MY), a rural site, 90 km northeast of the city center. It is near the Miyun Reservoir, and has no anthropogenic emissions within 5 km. PM_{10} samples were collected at the MY site during two collection periods, i.e., late spring (2011/05/02–2011/06/13) and summer (2011/07/28–2011/08/26). 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 an impactor with a cut-off aerodynamic diameter at 2.5 μm (G1200-41). Quartz filters (8 × 10 in, Pall Corporation, NY, USA), prebaked at 550 °C for at least 8 h, were used for sampling. Details of the sampling procedures were described in Liang et al. (2013a).

2.2. Sample analysis

Quartz filters were analyzed for OC by a thermal/optical carbon analyzer (DRI Model 2001, Desert Research Institute, USA), using the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal evolution protocol with reflectance charring correction. Briefly, a quartz filter punch (0.495 cm^2) was placed on a quartz boat inside the thermal desorption chamber of the analyzer, and then heated sequentially in He (to 580 °C) and He/O_2 (to 840 °C) atmosphere to volatilize and combust the loaded carbon, respectively (Cheng et al., 2014). The analytical error of OC was within 10%, and one sample of every 10 samples was selected at random for duplicate analysis. The OC detection limit was 0.82 μg cm^{-2}.

The quartz filter samples were also analyzed for a fungal tracer, i.e., mannitol, by high-performance anion exchange chromatography with pulsed amperometric detection (HPAEC-PAD) on a Dionex ICS-3000 system (Dionex, Sunnyvale, CA, USA). A punch (2.2 cm^2) of each quartz filter was extracted with 2.0 mL of deionized water (>18.2 MΩ resistivity) under ultrasonic agitation for 60 min, then filtrated through syringe filters (0.45 μm, Pall Corporation, NY, USA). Authentic standards for all carbohydrate species, including mannitol, were used for their quantification based on multi-point calibration curves. The extraction efficiency of this analytical method was determined to be better than 90% based on analysis of quartz filters spiked with known amounts of mannitol. More details about the HPAEC-PAD method can be found in Liang et al. (2013a).

2.3. Determination of fungal spore number concentration and carbon content

The average content of mannitol per spore (0.49 ± 0.20 pg) obtained at the same urban site in Beijing, China was applied in this study (Liang et al., 2013b). Combined with the year-round mass concentrations of mannitol, the number concentrations of fungal spores were obtained. The carbon content of fungal spores was calculated using a conversion factor of 13 pgC $spore^{-1}$ obtained earlier as the average carbon content of spores from nine airborne fungal species, with an uncertainty of 20% (Bauer et al., 2008b). Thus, using these conversion factors with analytical data for mannitol in filter samples, the contribution of fungal spores to the OC and to the mass balance of atmospheric aerosol particles can be estimated.

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