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Significant influence of fungi on coarse carbonaceous and potassium aerosols in a tropical rainforest

Zhisheng Zhang[1], Guenter Engling[2,3], Leiming Zhang[4], Kimitaka Kawamura[5], Yihong Yang[6], Jun Tao[1], Renjian Zhang[7], Chuen-yu Chan[8] and Yide Li[9]

1. South China Institute of Environmental Sciences, Ministry of Environmental Protection, Guangzhou, People's Republic of China
2. Department of Biomedical Engineering and Environmental Sciences, National Tsing Hua University, Hsinchu, Taiwan
3. Division of Atmospheric Sciences, Desert Research Institute, Reno, NV, USA
4. Air Quality Research Division, Science Technology Branch, Environment Canada, Toronto, Canada
5. Institute of Low Temperature Science, Hokkaido University, Sapporo, Japan
6. School of Chemical Engineering and Materials Science, Beijing Institute of Technology, Zhuhai, People's Republic of China
7. RCE-TEA, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, People's Republic of China
8. Key Laboratory of Aerosol, SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, People's Republic of China
9. Institute of Subtropical Forestry, Bureau of Forestry, Guangzhou, People's Republic of China

E-mail: zhangzhisheng@ scies.org and guenter@mx.nthu.edu.tw

Abstract

Fungal spores are ubiquitous in the Earth's atmosphere, especially in the environment of tropical rainforests with intense biological activities. To assess the impact of fungi on chemical components of atmospheric aerosols at a Chinese tropical rainforest site, size-segregated fungal spore tracers (i.e. arabitol and mannitol) were measured along with major aerosol components, including carbonaceous species and water-soluble inorganic ions. The fungal spore tracers were found to be predominately associated with coarse particles, in which organic carbon (OC) and potassium (K^+) were also present at significant levels. Enhanced amounts of fungal spore tracers were closely linked to rainfall events. Moreover, fungal spore tracers exhibited positive correlations with relative humidity and negative correlations with wind speed, temperature or radiation. The relationships between fungal spore tracers and meteorological factors are consistent with the emission features of actively discharged fungal spores, which are generally associated with sugar alcohols and by-products such as the inorganic ion K^+. The excellent correlations between fungal spore tracers and OC or K^+ in the coarse particles further suggested their common emission sources. Absolute principal factor analysis further identified fungi as the largest contributor to coarse OC and K^+ (both at ~66%) in this rainforest.

1. Introduction

Primary biological aerosol particles (PBAPs) are ubiquitous in the atmosphere, comprised of diverse types of biological substances such as fungal spores, pollen grains, bacteria and viruses (Jaenicke 2005, Pöschl 2005, Després *et al* 2012). In recent years, more attention has been paid to the influence of PBAPs on climate, atmospheric chemistry and physics, as well as public health, especially in areas where biological activities are intense (Després *et al* 2012).

Spores and associated materials released by fungi are the most abundant source of PBAPs in outdoor air, making up considerable fractions of atmospheric particulate matter (Elbert *et al* 2007, Bauer *et al* 2008b,

Zhang *et al* 2010). However, their abundance may vary highly with location, season, weather condition and land use. About 4% of PM_{10} and 8% of organic carbon (OC) were estimated to be from fungal spores at an urban and a suburban site in Vienna (Bauer *et al* 2008b). The contribution of fungal spores to PM_{10} could be up to ~35% in the Amazonian tropical rainforest (Elbert *et al* 2007). 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). In view of their role serving as cloud condensation nuclei and ice nuclei, as well as their long-range transport in the tropical region along with active atmospheric circulation (Pöschl

et al 2010), fungal aerosols may be particularly important in tropical rainforests.

The by-product components accompanied with PBAP emissions may play important roles in atmospheric physics and chemistry as well. For example, unexpected potassium salt particles were recently found to efficiently initiate the growth of organic aerosols in the Amazonian rainforest, where new particle formation was not often observed (Pöhlker *et al* 2012). Potassium particles can act as seeds for condensation of low or semi-volatile organic compounds, activating the formation of secondary organic aerosols (SOA). Pöhlker *et al* (2012) further suggested that plants and fungi may be the potential sources of potassium particles. However, to date there is still a lack of direct evidence to confirm the origins of potassium particles in forest areas.

The present study focuses on investigating the influence of fungi on atmospheric particulate components, especially OC and K^+, in a Chinese tropical rainforest, using fungal spore tracers (i.e. arabitol and mannitol). The polyols arabitol and mannitol are common sugar storage compounds of fungal spores in appreciable amounts and have been proposed as tracers to quantify the contribution of fungal spores to ambient aerosols (Bauer *et al* 2008a, Zhang *et al* 2010). We report size distribution patterns of fungal spore tracers, and discuss potential release mechanisms of fungal spores based on the relationships between tracers and meteorological factors. We further estimate the contributions of fungi to OC and K^+ using the absolute principal component analysis (APCA) method, to assess the influence of fungi on aerosol components.

2. Experimental

2.1. Sampling description

The sampling site (18°14′N, 108°49′E, 820 m A.S.L.) is located in the central part of the Jianfeng mountain (JFM) natural reserve on Hainan Island, China. The natural reserve has the largest and best-preserved tropical rainforest in China and is characterized by intense biological activity. It faces the South China Sea in the south and west directions, and is more than 120 km away from any major city. The sampling site is thus generally regarded as a remote site of South China with very limited local anthropogenic emission sources. More information about this site can be found in Tang *et al* (2009).

The sampling campaign was conducted during May and June 2010. A six-stage high-flow impactor (Model 131, MSP Corporation, Shoreview, MN, USA) was used to collect size-segregated aerosols. The impactor was equipped with a pre-impaction stage and six regular stages with cut-point diameters of 18, 10, 2.5, 1.4, 1.0, 0.44 and 0.25 μm, respectively. However, results obtained from the pre-impaction stage

(i.e. size between 10 and 18 μm) were not included in the discussion considering its low aerosol mass loading. The high-flow impactor loaded with pre-baked quartz filters was operated at a flow rate of 100 L min^{-1} for approximately 48 h to collect a set of size-segregated aerosol samples. Blanks followed the entire procedure as ambient sample collection, except with the pump being turned off. The collected samples were stored at <4 °C before chemical analysis. Twenty sets of aerosol samples and three sets of blanks were collected during the campaign. Meteorological parameters including rainfall amount, relative humidity (RH), temperature, radiation, and wind speed were also measured at a weather station about 50 meters away from the sampling site.

2.2. Chemical analysis

Sugar alcohols (i.e., arabitol and mannitol) and other carbohydrates including levoglucosan were determined by high-performance anion-exchange chromatography with pulsed amperometric detection, which is a well-established method for poly-hydroxy organic compound measurement. The detailed analysis procedures can be found in Zhang *et al* (2013). Briefly, a quarter of each filter was extracted with deionized ultra-pure water in pre-baked glass bottles under ultrasonic agitation for 60 min. The extracts were filtered through Teflon syringe filters to remove insoluble materials, and were subsequently analyzed on a Dionex ICS-3000 system with a Dionex Carbopac MA1 analytical column. The limit of detection (LOD) of both arabitol and mannitol was estimated to be 0.2 ng m^{-3} in this study. The anhydrosugar and polyol contents in all field blanks were lower than the LOD.

A portion of each quartz filter (1.5 cm^2) was cut and used to determine the carbonaceous species using a thermal/optical transmittance aerosol carbon analyzer (Sunset Laboratory, OR, USA). The first stage of carbon analysis was performed in an inert helium atmosphere, while the second stage was conducted under an environment of 2% O$_2$ /98% He. The detailed protocol employed here can be found in Lin *et al* (2014). It should be noted that OC reported here refers to the carbon content detected in the first stage, while EC corresponds to the carbon content measured in the second stage. This is because the laser correction did not work properly to separate OC and EC due to the non-uniform particle deposition on the filters collected by the cascade impactor. A similar definition of carbonaceous species has also been applied previously (Huang and Yu 2008). Blank samples were analyzed according to the same procedures as the ambient samples, and the results of all the ambient samples reported here have been blank corrected. The LODs of OC and EC were both estimated to be 0.02 μgC m^{-3}.

Water-soluble ions were quantified in the same extracts as for carbohydrate analysis on a Dionex ICS-3000 ion chromatograph. The major inorganic cations

Table 1. Statistics of size-segregated mass concentrations of fungal spore tracers, carbonaceous species and water-soluble potassium.

Species	2.5–10 μm	1.4–2.5 μm	1.0–1.4 μm	0.44–1.0 μm	0.25–0.44 μm	<0.25 μm
Arabitol (ng m^{-3})	53.1 ± 30 (20)	7.0 ± 2.4 (20)	0.4 ± 0.1 (15)	0.4 ± 0.1 (13)	0.4 ± 0.2 (5)	0.3 ± 0.1 (3)
Mannitol (ng m^{-3})	75.1 ± 44 (20)	8.0 ± 2.7 (20)	0.4 ± 0.3 (15)	0.4 ± 0.2 (9)	0.4 ± 0.2 (2)	BDL
OC (ngC m^{-3})	923 ± 382 (18)	214 ± 32 (18)	147 ± 39 (18)	546 ± 222 (18)	423 ± 204 (18)	648 ± 211 (18)
EC (ngC m^{-3})	140 ± 85 (18)	50 ± 35 (18)	34 ± 27 (18)	147 ± 86 (18)	97 ± 49 (18)	117 ± 82 (18)
TC (ngC m^{-3})	1063 ± 447 (18)	264 ± 49 (18)	180 ± 59 (18)	693 ± 250 (18)	520 ± 228 (18)	765 ± 283 (18)
K$^+$ (ng m^{-3})	30.8 ± 14.8 (20)	10.0 ± 5.5 (20)	10.3 ± 8.1 (20)	51.7 ± 22.1 (20)	30.2 ± 14.0 (20)	10.4 ± 7.5 (20)

Note: the data is presented as mean ± one standard deviation, and the values in the parentheses are the number of samples above the detection limit. Only samples with values above the detection limit are used for calculating mean and standard deviation. BDL in the table represents all samples being below detection limit.

(K^+, Na^+, Mg^{2+}, Ca^{2+}, and NH_4^+) were separated on an Ionpac CS12 analytical column with CG12 guard column using 20 mM methanesulfouic acid as eluent at a flow rate of 1.0 mL min^{-1}. The major anions (SO_4^{2-}, NO_3^- and Cl^-) were separated on Ionpac AS14 and AG14 columns with a mixture of Na_2CO_3 and $NaHCO_3$ as eluent at a flow rate of 1.2 mL min^{-1}. The LOD of K^+ was estimated to be 0.8 μg m^{-3}.

2.3. Statistical analysis

To investigate the influence of fungi on organic and inorganic aerosols, a well-developed multivariate receptor model (i.e., APCA) was applied for source identification and apportionment. The original data set was first used to transform the observed particulate species into a set of uncorrelated variables using varimax rotated PCA. The absolute principal component scores (APCS) for each sample were subsequently calculated. Lastly, source contributions of each principal component (PC) to the target species were obtained using stepwise multiple regression of particulate species in the APCS. A detailed description of this method can be found in Thurston and Spengler (1985). Statistical analysis of data such as correlation analysis and PCA were conducted using SPSS statistical software.

3. Results and discussion

3.1. Size distributions of sugar alcohols, carbonaceous species and potassium

Statistical results of size-segregated fungal spore tracers, carbonaceous aerosol components, and water-soluble potassium observed at the JFM site are listed in table 1, and their average mass size distribution patterns are shown in figure 1. Arabitol and mannitol were mostly present in particle size ranges of 1.4–2.5 μm and 2.5–10 μm (referred to as coarse particles hereafter) with negligible amounts in smaller particles. The fractions of arabitol and mannitol in the coarse mode accounted for 85% and 88%, respectively, of their respective total mass concentrations. The size distributions of arabitol and mannitol are consistent with the size range of airborne fungal spores reported in previous studies, which showed

aerodynamic diameters typically ranging from 2 to 10 μm, although a few fungal spores could exceed 10 μm (Elbert *et al* 2007, Huffman *et al* 2012).

Total carbon (TC) was mainly composed of OC with only a small contribution from EC. As shown in figure 1, the size distribution of OC was characterized by a bimodal pattern with one peak in the coarse mode and the other in the fine mode. TC had a similar size distribution due to the dominance of OC in TC. The EC size distribution, as expected, was characterized by a unimodal pattern peaking at the fine particle size. The OC/EC ratio is often applied to examine the sources of carbonaceous aerosols and to evaluate SOA contributions (Turpin and Huntzicker 1995). The average OC/EC ratio ranged from 4.7 to 7.8, depending on particle size, with a maximum observed in the coarse mode. These values are substantially higher than those (~1.5) observed in many Chinese urban locations (Cao *et al* 2007). The characteristic low EC level and high OC/EC ratio observed in this forest area can be explained by the extensive biogenic carbon aerosol emissions and/or intensive biogenic SOA formation.

A unimodal size distribution was found for water-soluble K^+ peaking in the fine mode, while appreciable amounts of K^+ were also found in the coarse mode (see table 1). This finding was somewhat different from that observed in the Amazonian rainforest, where K^+ was mostly concentrated in coarse particles (Graham *et al* 2003). However, the average coarse K^+ concentration obtained here was indeed comparable to that reported for the Amazonian rainforest. The high abundance of K^+ in fine particles in this study was likely a result of long-range transport of biomass burning aerosols, noting that they have a longer lifetime than coarse particles. It has been reported that biomass burning smoke plumes originating from Southeast Asia and the Philippines can be transported to coastal South China, resulting in elevated PM$_{2.5}$ and K^+ concentrations (Zhang *et al* 2012). This phenomenon was also observed in this study in one sample collected during May 8–10 when the highest K^+ and levoglucosan concentrations in fine particles were detected, but with no obvious increase of coarse K^+ and fungal spore tracers.
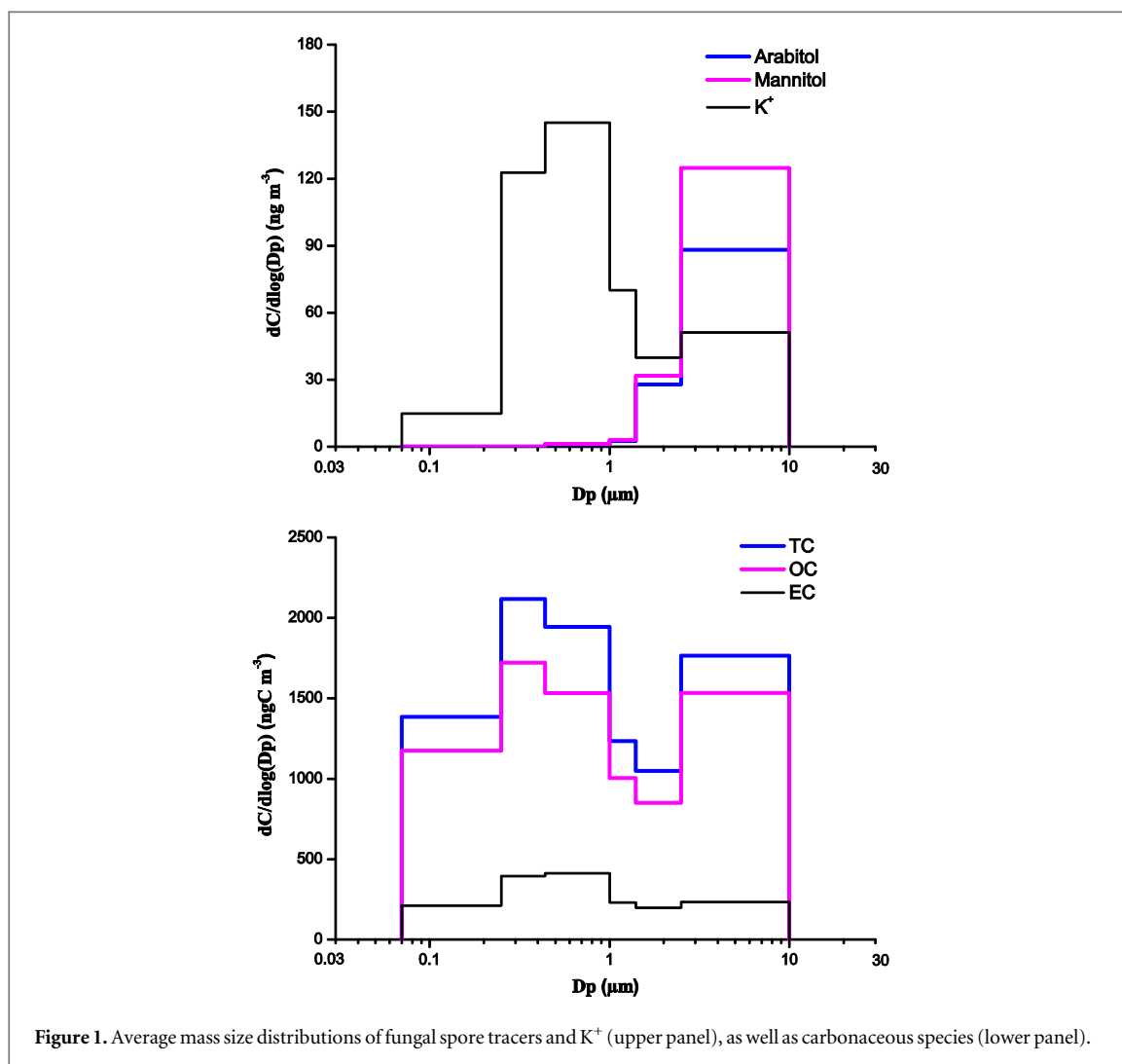
Figure 1. Average mass size distributions of fungal spore tracers and K$^+$ (upper panel), as well as carbonaceous species (lower panel).
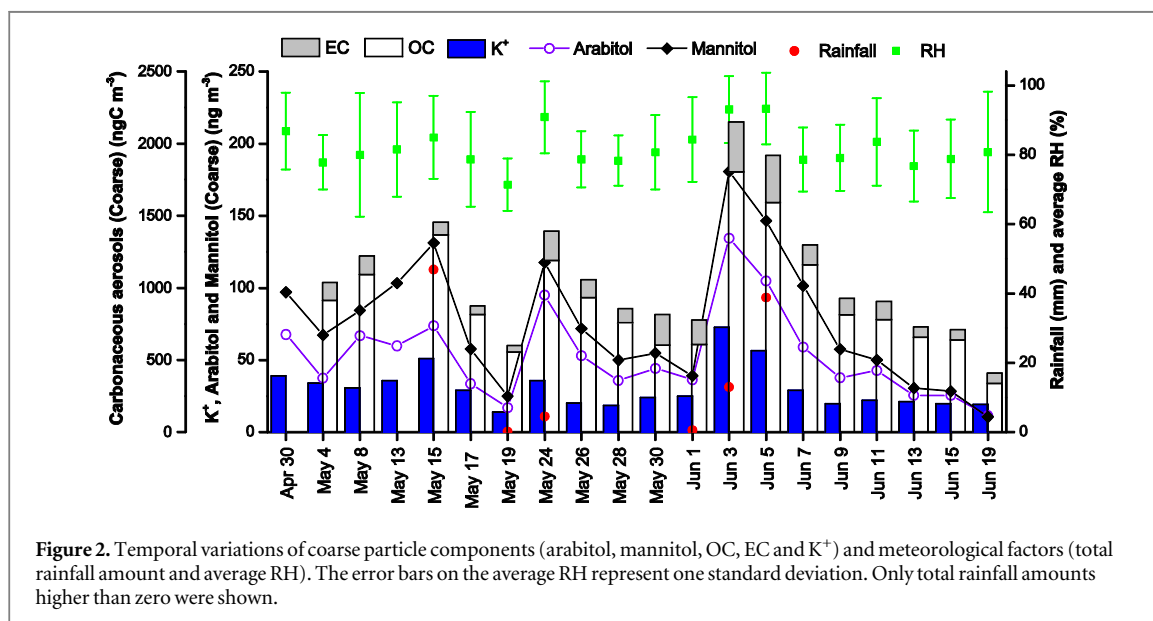
Figure 2. Temporal variations of coarse particle components (arabitol, mannitol, OC, EC and K$^+$) and meteorological factors (total rainfall amount and average RH). The error bars on the average RH represent one standard deviation. Only total rainfall amounts higher than zero were shown.

3.2. Influence of meteorological factors on the distribution of fungal spore tracers

Meteorological factors can significantly influence the initial release of fungal spores and affect their subsequent dispersal in the air (Jones and Harrison 2004). The relationships between fungal spore tracers and meteorological factors were examined here to investigate the possible sources and release

Table 2. Pearson coefficients between coarse-mode fungal spore tracers and meteorological factors at the JFM site ($n = 20$).

	Air pressure	Radiation	Wind speed	Temperature	RH
Arabitol	0.159	−0.566[a]	−0.663[b]	−0.841[b]	0.847[b]
Mannitol	0.204	−0.477[a]	−0.521[b]	−0.847[b]	0.764[b]

Note:

[a] Represents correlation is significant at the 0.01 level (2 tailed).

[b] Represents correlation is significant at the 0.001 level.

mechanisms of airborne fungal spores. The temporal variations of coarse arabitol and mannitol throughout the campaign are exhibited in figure 2. It is clearly shown that the increases in fungal spore tracer levels were consistently associated with rainfall events, although the correlation between tracer concentrations and rainfall intensities was poor. The release of fungal spores appeared to be stimulated by precipitation, as was also observed at a forest site in the moderate climate zone (Huffman *et al* 2013). In that study it was observed that biological particle concentrations were dramatically increased during and after rain, which even surpassed wet scavenging effects. During this sampling campaign, ambient temperatures ranged between 18 and 31 °C, and RH was consistently higher than 51% (with an average of 82%). Such a warm and humid environment was conducive for the growth and reproduction of fungi. Table 2 shows the correlation coefficients between fungal spore tracer concentrations and meteorological factors. Coarse arabitol or mannitol exhibited a positive correlation with RH and a negative correlation with wind speed, temperature or radiation, indicating enhanced airborne fungal spore levels under wet, cool and calm ambient conditions. The relationships between fungal spore tracers and meteorological factors observed here are consistent with those from our previous study conducted at JFM (Zhang *et al* 2010). However, it was found that a higher abundance of fungal spore tracers in an urban area of Beijing occurred at moderate RH levels (51–70%) (Liang *et al* 2013), which might be due to the difference in fungi species dominating in different environments.

The spore release of major fungi classes can be generally classified into active and passive discharging mechanisms. Active discharging frequently requires a supply of water from the surrounding environment to eject spores by means of osmotic pressure and surface tension effects (Elbert *et al* 2007). Most ascospores and basidiospores are known to be released by the active discharging mechanism. Precipitation appears to be required for ascospore release, as enhanced ascospore concentrations are frequently observed during and after rainfall (Gottwald *et al* 1997). Many basidiospores, on the other hand, appear to be more correlated with RH (Burch and Levetin 2002). In contrast, the passively discharged spores depend on air movement or other external forces. *Cladosporium* spores, for

example, have been shown to be positively correlated with temperature and negatively correlated with RH and rainfall level (Oliveira *et al* 2009). Jones and Harrison (2004) suggested that the increase in temperature and decrease in RH can reduce the binding effect of passively discharged spores to fungi, and strong wind is helpful for the detachment and the dispersal.

Therefore, actively discharged spores tend to be increased under wet and humid conditions, while passively discharged spores tend to be enhanced under dry, warm and windy weather conditions. Consequently, we can expect that the findings in this study are indications of actively discharged spores. This is consistent with the observation reported for the Amazonian rainforest, where the presence of atmospheric fungi was dominated by actively discharged spores (Elbert *et al* 2007).

3.3. Contributions of fungi to coarse carbonaceous and potassium aerosols

The temporal variations of coarse OC (or TC) and K^+ concentrations were very similar to those of coarse fungal spore tracers. It has been pointed out that active release of fungal spores is probably accompanied by emission of aqueous droplets containing carbohydrates and inorganic ions such as potassium (Elbert *et al* 2007). The excellent correlations between coarse OC or K^+ and fungal spore tracers (figure 3) indicated that coarse OC and K^+ have common sources which are related to fungal activity.

To assess the contribution of fungi to OC and K^+, aerosol components measured in coarse and 1.4–2.5 μm size particles were analyzed using PCA since considerable amounts of fungal spore tracers were detected in these particle size ranges. Table 3 displays the input species and the varimax rotated PC loading matrix results. Four PCs were extracted with eigenvalues greater than unity, accounting for 92.9% of the total variance in the data set. The communalities for all of the species were mostly greater than 85% except for calcium (77%). PC1 was characterized by high loading of arabitol, mannitol, carbonaceous species and potassium, which appeared to represent fungal aerosols. PC2 was interpreted as sea-salt aerosols and secondary nitrate since it is highly correlated with the sea salt components (i.e. Na^+, Cl^- and Mg^{2+}) and nitrate formed upon sea-salt aerosols. PC3 mostly depended on sulfate and ammonium and thus can be
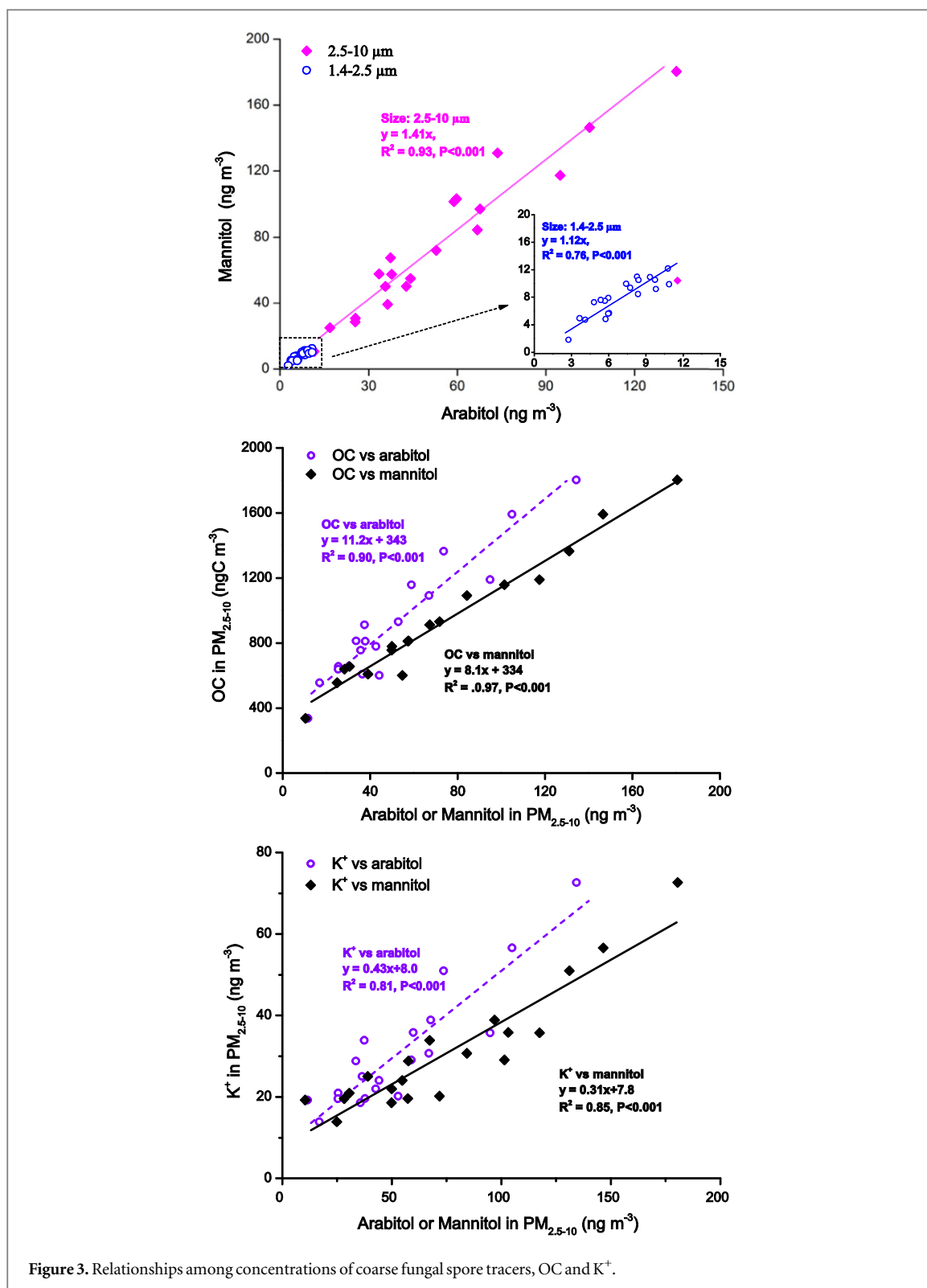
Figure 3. Relationships among concentrations of coarse fungal spore tracers, OC and K^+.

regarded as secondary ammonium sulfate aerosols. The interpretation of PC4 was biomass burning and dust aerosols based upon its rather high loading with levoglucosan and calcium.

The contribution of each PC group to OC and K^+ were then quantitatively assessed by means of stepwise multiple linear regression. The multiple linear regression was applied by employing the target species (OC, K^+, arabitol and mannitol) as dependent variables and the APCS obtained from the PCA as independent variables. The resultant regression coefficients were then used to convert the APCS for each sample to produce the estimates of each PC contribution, while the regression constant was regarded as the contribution from the non-specified sources. The multiple linear regression of target species on APCS exhibited

Table 3. Varimax rotated principal component loading matrix.

Species	PC 1	PC 2	PC 3	PC 4	Communalities
Arabitol	0.98	0.12	−0.07	−0.01	0.99
Mannitol	0.96	0.22	−0.05	−0.07	0.98
OC	0.94	0.26	−0.09	−0.01	0.96
K^+	0.92	0.28	0.22	0.02	0.97
EC	0.92	−0.07	0.22	0.20	0.94
Na^+	0.06	0.96	−0.05	−0.06	0.92
Mg^{2+}	0.22	0.95	−0.07	0.13	0.97
Cl^-	0.11	0.94	0.00	−0.07	0.91
NO_3^-	0.30	0.84	−0.13	0.30	0.91
SO_4^{2-}	0.10	0.06	0.97	0.03	0.95
NH_4^+	0.00	−0.22	0.95	0.01	0.94
Levoglucosan	−0.12	−0.06	0.05	0.91	0.85
Ca^{2+}	0.34	0.46	−0.02	0.67	0.77
Eigenvalue	6.02	2.93	1.82	1.31	
% Of variance	46.3	22.5	14.0	10.1	
Potential sources	Fungi related aerosols	Sea-salt and secondary nitrate	Secondary sulfate and ammonium	Biomass burning and dust aerosols	

Note: Only aerosol components measured in the particle sizes of 2.5–10 and 1.4–2.5 μm served as input in principal component analysis.

excellent agreement between predicted and observed values with the correlation coefficient higher than 0.95 and the slope close to 1.

Mean source contributions of PCs to target species for the coarse and 1.4–2.5 μm size particles are shown in figure 4. As expected, arabitol and mannitol were almost exclusively present in the fungi related aerosol factor. The largest fractions of OC and K^+ were both assigned to PC1, especially in the coarse particles where 66.4% of OC and 65.4% of K^+ on average were associated with this factor. The mean contributions of PC1 dropped down to around 42% for both species in the 1.4–2.5 μm size particles, as the influence of fungi became weaker. Above 20% of OC and K^+ were apportioned to the sea salt factor. Organic aerosols in forest regions typically consist of a significant amount of biogenic SOA, formed from the photo-oxidation of the biogenic volatile organic compounds and condensation of low- and semi-volatile oxidation products. Pre-existing sea salt particles could act as seeds for SOA formation (Beardsley *et al* 2013). It was not surprising that a considerable amount of OC was associated with sea salt aerosols. As for K^+, it is a major chemical component of sea salt. The sea salt potassium (SS-K^+), using magnesium as sea salt reference (SS-K^+ = [K^+] −0.159*[Mg^{2+}], Cheng *et al* 2000), accounted for 17.1% and 17.8% of the total potassium in coarse and 1.4–2.5 μm size particles, respectively. The results were in good agreement with those obtained from APCA.
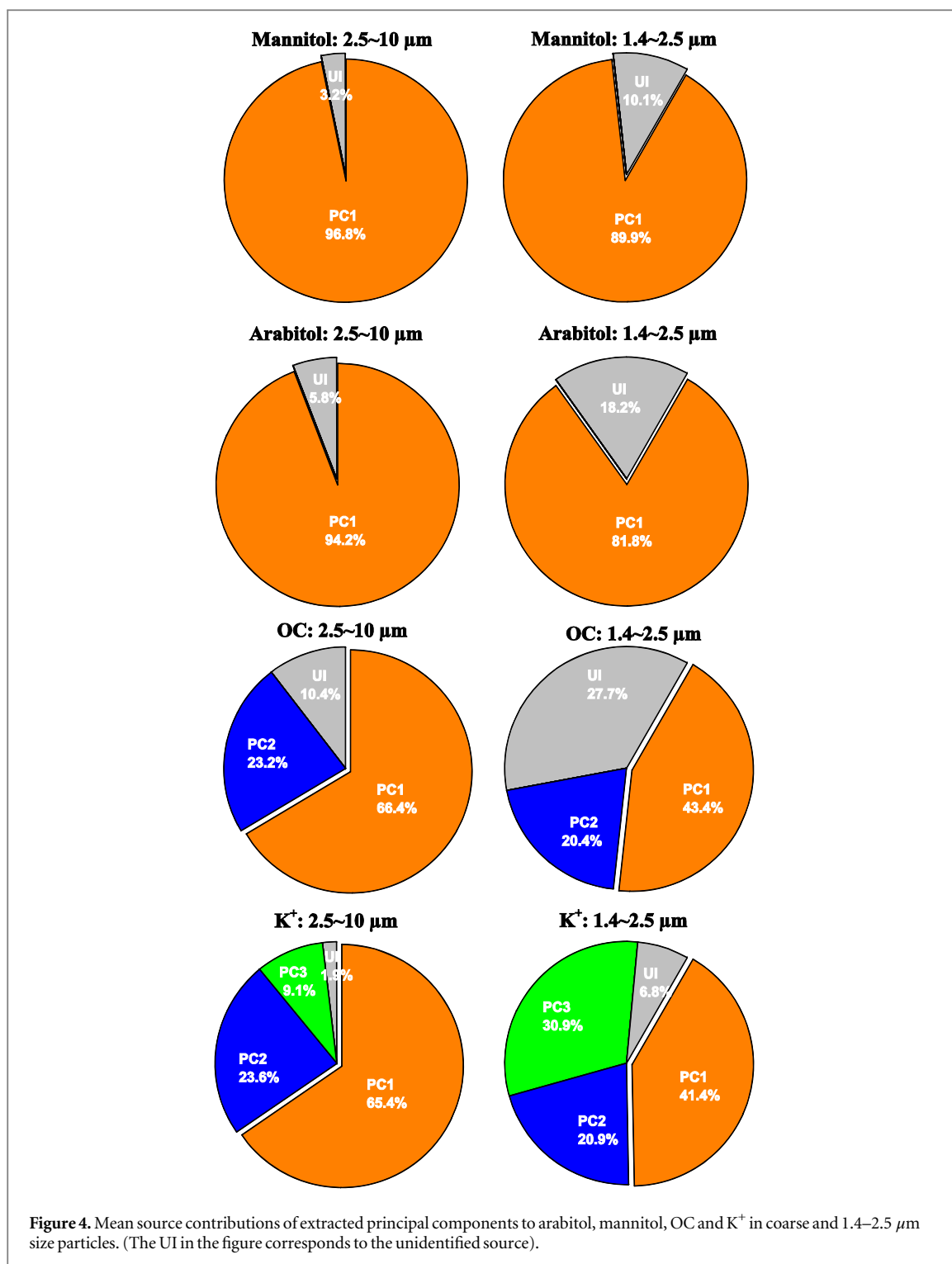
An experimentally determined conversion factor method was previously applied to estimate the contribution of fungal spores to OC (Bauer *et al* 2008b). Reported conversion factors for estimating spore numbers from arabitol mass ranged from 0.8 to 1.8 (mean: 1.2) pg arabitol spore^{-1}, and from mannitol mass ranged from 1.2 to 2.4 (mean:1.7) pg mannitol

spore^{-1}. If the range and mean values of these conversion factors were used, total spore number concentrations during the present campaign were estimated to range from 33×10^3 to 75×10^3 m^{-3} with an average of 50×10^3 m^{-3}. Reported common airborne spore carbon contents ranged from 5.2 to 23 (mean: 13) pg carbon spore^{-1} (Bauer *et al* 2002). Using these numbers, spore-derived OC was estimated to range from 434 to 1734 ngC m^{-3} with an average of 651 ngC m^{-3}. The estimated spore-derived OC is 38%–151% (mean: 57%) of the observed coarse OC, suggesting significant contributions of fungi sources to ambient coarse OC. The large range of the above estimated numbers also suggests the necessity of more field studies, considering the site-dependence of the conversion factors. Critical meteorological and biological factors controlling spore release need to be first identified before more accurate methods can be developed to quantitatively assess fungal sources of OC.

4. Conclusions

Size-segregated fungal spore tracers and major aerosol components were measured at a tropical rainforest site in South China. Arabitol and mannitol were exclusively present in the size range of 1.4–10 μm, indicating fungal spores were mainly associated with coarse particles. The size distribution of OC exhibited a bimodal distribution with one mode being in the fine and the other in the coarse size range. The size distribution of K^+ was characterized by a unimodal pattern but in appreciable concentrations in both fine and coarse particles.

The coarse arabitol and mannitol concentrations highly correlated with RH at the forest site, and were closely linked to precipitation, indicating actively

Figure 4. Mean source contributions of extracted principal components to arabitol, mannitol, OC and K$^+$ in coarse and 1.4–2.5 μm size particles. (The UI in the figure corresponds to the unidentified source).

discharged spores. Correlation analysis between the coarse fungal spore tracers and coarse OC or K$^+$ suggested that they were likely from common sources. As by-products of actively discharged spore emissions, organic matter and potassium may be co-emitted into the air. The contributions of fungi to coarse OC or K$^+$ were estimated to be as high as 66% on average at the JFM site using the APCA. Similar results can also be obtained if applying a simplified conversion factor method. It is thus concluded that fungi are not only

the major source of coarse OC, but also the most important source for coarse K$^+$ in tropical forests. More studies are needed to develop methods for quantitatively estimating the contributions of fungi to coarse OC and K$^+$ particles.

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