Patterns in airborne pollen and other primary biological aerosol particles (PBAP), and their contribution to aerosol mass and number in a boreal forest

Hanna E. Manninen[1)2)]*, Jaana Bäck[3)], Sanna-Liisa Sihto-Nissilä[1)],
J. Alex Huffman[4)5)], Anna-Mari Pessi[6)], Veijo Hiltunen[1)], Pasi P. Aalto[1)],
Pablo J. Hidalgo[7)], Pertti Hari[3)], Annika Saarto[6)], Markku Kulmala[1)] &
Tuukka Petäjä[1)]

[1)] *Department of Physics, P.O. Box 64, FI-00014 University of Helsinki, Finland (*corresponding author's e-mail: hanna.manninen@helsinki.fi)*
[2)] *Institute of Physics, University of Tartu, Ülikooli 18, EE-50090 Tartu, Estonia*
[3)] *Department of Forest Sciences, P.O. Box 27, FI-00014 University of Helsinki, Finland*
[4)] *Department of Chemistry and Biochemistry, University of Denver, 2190 E. Illif Ave., Denver, CO 80208, USA*
[5)] *Max Planck Institute for Chemistry, P.O. Box 3060, D-55020 Mainz, Germany*
[6)] *Department of Biology, FI-20014 University of Turku, Finland*
[7)] *Department of Environmental Biology, University of Huelva, ES-21071 Huelva, Spain*

We studied variation in concentrations of airborne pollen and other particles of biological origin in a boreal forest in Finland during 2003–2004. The highest concentrations of pollen were observed in late spring and early summer, whereas the peak concentrations of other particles of biological origin (including e.g. fungal spores) occurred in August–September. Although the patterns in concentrations in 2003 and 2004 were similar, the concentration levels were significantly different between the years. The contribution of pollen and other particles of biological origin led to an increase in the measured particulate matter (PM) mass during the pollen season (mass of pollen and other particles of biological origin 5.9 and 0.4 μg m^{-3}, respectively, in respect to PM$_{total}$ mass of 9.9 μg m^{-3}) but the effect on total particle number was negligible. The other particles of biological origin constituted the largest fraction of measured primary biological aerosol particle (PBAP) numbers (~99%), whereas pollen showed a higher relative mass fraction (~97%) of PBAP. These results underline the important contribution of PBAP to coarse atmospheric particle mass providing up to 65% of the total mass during the peak pollen season.

Introduction

Primary biological aerosol particles (PBAP), or

bioaerosols, are emitted from vegetation and by other living organisms. PBAP include pollen, fungal spores, bacteria, viruses, cell fragments,

and protozoans (Després *et al.* 2012) and they are ubiquitous in the atmosphere (Gregory 1961, Womack *et al.* 2010). The main research interest regarding PBAP have been directed to their effects on humans, animals and agriculture (e.g. Waggoner 1983, Burge 1990), their potential as agents of biological warfare (e.g. Lim *et al.* 2005), and the environmental processes they contribute to, e.g. ice and liquid water cloud droplet activation (Després *et al.* 2012, Morris *et al.* 2013), and atmospheric chemistry (Deguillaume *et al.* 2008, Vaitilingom *et al.* 2013). Pollen and fungal spores cause allergic symptoms in humans (Schappi *et al.* 1999, Barnes *et al.* 2000, Simon-Nobbe *et al.* 2008), in particular during spring when pollen concentrations are typically the highest.

Recently, there has been growing interest in studying the impact of bioaerosols on cloud formation and precipitation (Möhler *et al.* 2007, Pöschl *et al.* 2010, DeMott *et al.* 2011, Morris *et al.* 2011), and bacteria, spores and pollen have been introduced to global climate models as sources of primary particles (Heald and Spracklen 2009, Hoose *et al.* 2010, Spracklen *et al.* 2010, Sesartic *et al.* 2013). Fungal spores, pollen grains, and their fragments have been shown to nucleate ice at relatively high temperatures in the laboratory, suggesting that these particle classes may contribute to atmospheric cloud formation and evolution if lofted in sufficient numbers (e.g. Diehl *et al.* 2001, Pummer *et al.* 2012, Haga *et al.* 2013), and *in situ* measurements at ground level and in clouds at high altitude have corroborated this possibility (e.g. Prenni *et al.* 2009, DeLeon-Rodriguez *et al.* 2013, Huffman *et al.* 2013, Tobo *et al.* 2013). There are indications that biological particles could be important for the cloud water cycle especially in boreal forest region (Morris *et al.* 2013, Sesartic *et al.* 2013). As bioaerosols contribute significantly to particle mass (Jaenicke *et al.* 2007, Pöschl *et al.* 2010), it is clear that biological aerosol particles exert a potentially large impact on the climate system. In view of a globally changing climate, it is possible that the effect of biological particles will be even more pronounced in the future, as the growing seasons get longer (Beggs 2004, Weber 2012).

Individual pollen grains can range from approximately 10 to 100 μm in diameter, whereas fungal spores are typically smaller, 1–50 μm, most commonly < 10 μm (Després *et al.* 2012). Bioaerosols constitute a relevant fraction of aerosol particles in both accumulation and coarse modes; it is estimated that globally they constitute ~25% of aerosol particles in the size range 0.2–50 μm both by mass and number (Jaenicke 2005). In most areas, the fraction of biological aerosol has been reported to be 15%–25%, but in the Amazonian region biological particles contribute up to 85% of coarse particle mass (Jaenicke *et al.* 2007, Pöschl *et al.* 2010). There is on-going debate on the source strength estimates of bioaerosols. Some studies (Jaenicke *et al.* 2005, Elbert *et al.* 2007) suggest that bioaerosol emission factors within current climate models are at the moment underestimated. To get reliable estimates on emission rates at global and annual scales, there is an urgent need for long-term measurement data on concentration of primary biological aerosol particles.

In the boreal forest, most of the native tree species are wind-pollinated and thus high pollen concentrations can be expected in favourable conditions. In the boreal region, the highest concentrations of pollen in the atmosphere are recorded in late spring and early summer (Siljamo *et al.* 2008, Yli-Panula *et al.* 2009). Although the annual pattern of pollen appearance is similar from year to year, their concentration levels can change significantly depending on meteorological conditions during the growing season (Jones and Harrison 2004). Day-to-day variation in the PBAP concentrations is also large, temperature and humidity being the most important factors (Schumacher *et al.* 2013). Despite their large size, bioaerosols, including pollen grains, are aerodynamically buoyant (Reponen *et al.* 2001, Després *et al.* 2012) and can be transported over long distances via wind dispersal (Gregory 1978, Hjelmroos 1991, Prospero 2005, Sofiev *et al.* 2006, Hussein *et al.* 2013, Tack *et al.* 2014). Atmospheric transport of bioaerosols depends on prevailing meteorological conditions, such as wind speed, temperature, humidity, air turbulence and convection, and the physical size of the spores themselves (Madelin 1994, Jones and Harrison 2004, Norros *et al.* 2014). Although there is abundant information on seasonal patterns in concentrations of

pollen and other particles of biological origin, parallel field measurements of the above along with other airborne particles are scarce (Huffman *et al*. 2012, Schumacher *et al*. 2013).

The goal of this study was to analyse the temporal patterns in (1) airborne pollen grain concentrations, (2) concentrations of other particles of biological origin, and (3) aerosol particle mass and number concentrations in a boreal forest based on long-term *in situ* measurements. Further, the relationships between their concentrations and basic meteorological parameters were studied.

Particles of biological origin most commonly present in boreal forests, were collected by a Hirst-type spore sampler during two pollen seasons between March 2003 and September 2004, at the SMEAR II station in Hyytiälä in southern Finland. The number concentrations of PBAP are compared with real-time measurements of fluorescent bioaerosols and with total particle mass and number size distributions measured during the same period to evaluate the relative contribution of PBAP to the total particle matter (PM) mass.

Material and methods

Boreal forest site description

The measurements were performed at the SMEAR II (Station for Measuring Forest Eco-system–Atmosphere Relations II) station located in Hyytiälä, southern Finland (61°51´N, 24°17´E, 181 m a.s.l.). The station is equipped with extensive facilities to continuously and comprehensively measure forest-ecosystem–atmosphere interactions (Kulmala *et al*. 2001, Hari and Kulmala 2005). Hyytiälä is a rural background site with low local particle matter emissions. The station is surrounded by a managed Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*) dominated evergreen boreal forest, with a scattered mix of: willow (*Salix* spp.), birches (*Betula pendula* and *B. pubescens*), alders (*Alnus incana* and *A. glutinosa*), mountain ash (*Sorbus aucuparia*), common juniper (*Juniperus communis*) and trembling aspen (*Populus tremula*). Coniferous and mixed-conif-

erous-deciduous forests account for over 65% of the region surrounding Hyytiälä within a 50-km radius (Williams *et al*. 2011). Some information on the fungal spores in the Hyytiälä surroundings can be found in Hussein *et al*. (2013). Haapanala *et al*. (2007), Ilvesniemi *et al*. (2009) and Williams *et al*. (2011) provided detailed description of the land use and vegetation in the Hyytiälä surroundings (Fig. 1).

PBAP collection and identification

For the analyses reported here, all the PBAP data were pooled into two groups: (1) pollen and (2) other particles of biological origin (e.g., fungal and other spores, algae, nematoda and undefined species) as the data were too scarce to study all the species separately. Pollen were categorized into 15 vascular plant taxa. Other particles of biological origin were classified as spores of Deuteromycota, Basidiomycota, Ascomycota, Myxomycota, Bryopsida/Pteridophyta and *Equisetum*, or parts of mycelia (Appendices 1–4). These represent a majority of pollen and spore categories found in a boreal forest in southern Finland (Ranta and Satri 2007). Also some algae and nematoda were occasionally observed.

The pollen samples cover the periods from 7 March to 29 August 2003 and from 3 March to 30 September 2004 including the complete pollen seasons in both years. The samples of other particles of biological origin cover the periods from 6 March to 31 December 2003 and from 3 March to 4 August 2004. As in 2004 the sampling ended already in August, the 2004 samples do not include the complete season for other particles of biological origin.

The samples were collected using a Hirst-type volumetric spore trap (Burkard Manufacturing Co. Ltd.; Hirst 1952) henceforth called 'Burkard', which is the European standard sampling method for airborne pollen and spores (EAN, European Aeroallergen Network). The particles are collected on an adhesive, transparent, plastic tape (Melinex) mounted on a clockwork-driven drum. The trap has a built-in vacuum pump (10 l min⁻¹), designed to sample airborne particles larger than ~3 μm continuously for seven days. The tape is replaced
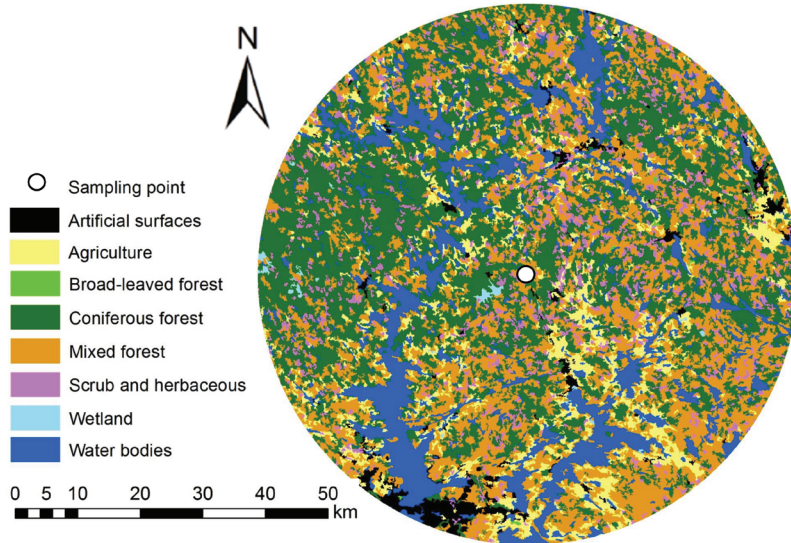
Fig. 1. Land-use map extracted from the Corine database for 50 km radius around the Hyytiälä forest site (EEA 1997, Williams *et al.* 2011).

weekly as the drum turns a full circle. The instrument is described in more detail in e.g. Battarbee *et al.* (1997). The trap was located 20 m above ground and approximately 3 m above the top of the canopy. The final PBAP concentration data have 24-hour time resolution. Sampling was carried out as described in Käpylä (1981), and counting and identification were performed according to standard methodology adopted by the Finnish pollen information network and following the principles of the European Aeroallergen Network (www.polleninfo.org/) and Rantio-Lehtimäki *et al.* (1994). The Burkard samples were examined by light microscopy (Smith *et al.* 2009), where the counting method was a stratified random sampling of microscopic fields, analysing 2.17% of total sampling area. The counts were normalised for the counted surface area and the number was reported as the number of grains per category and per volume of air (m^{-3}) (Mäkinen 1981).

Previously, Schumacher *et al.* (2013) reported year-round measurements of PBAP in Hyytiälä, using a laser-induced-fluorescence (LIF) method and a portion of these data is shown for comparison here. They used an ultraviolet aerodynamic particle sizer (UV-APS Model 3314, TSI Inc., Shoreview, MN, USA) which draws in ambient aerosol and measures aerosol number concentration classified according to their aerodynamic diameter between 1 and 20 μm (Huffman *et* *al.* 2010). A pulsed UV laser (355 nm) causes fluorescence of individual particles, which is detected in the wavelength range of 420–575 nm (single channel, not wavelength-dispersed). The LIF method provides a number concentration of fluorescent biological aerosol particles (FBAP) as a real-time proxy for PBAP, monitored at high time and size resolutions. However, biological particles which do not absorb laser light or whose fluorescence is weak, cannot be detected, and are typically undercounted by this technique (Pöhlker *et al.* 2012, 2013). With this method, bioaerosols cannot be classified to the species level without complementary techniques.

Physical characterization of aerosol particles

The particulate matter mass was measured in different size ranges with a Dekati PM10-impactor with gravimetric off-line analysis. The cascade impactor samples particles into four size ranges according to their aerodynamic particle size (PM$_1$: < 1 μm, PM$_{1-2.5}$: 1–2.5 μm, PM$_{2.5-10}$: 2.5–10 μm, PM$_{>10}$: > 10 μm and PM$_{total}$: sum of all PM). Polycarbonate membranes with no holes (25 mm diameter, Nuclepore® 800-203) were used as collection substrates in the first three stages of the impactor (largest particles). To prevent particle bouncing from the collection substrates,

membranes were lightly greased (Apiezon L). A Teflon® filter (47 mm diameter, 2 μm pore size, Gelman Teflo R2P J047) was used for collection of PM_1 mass in the last stage (smallest particles). The gravimetric analysis was performed with a Mettler Toledo MT5 microbalance. More detailed description of the instruments is given in Laakso *et al*. (2003). The impactor samples were collected continuously for 2–3 days at the time from a sample inlet 2 meters above ground (within the canopy).

Particle number and mass size distributions were measured in real time with an Aerodynamic Particle Sizer (APS, TSI Model 3321, TSI Inc., St. Paul, MN, USA) in the aerodynamic size range 0.5–20 μm. The APS inlet was mounted vertically, approximately 1.5 m above the roof of the building, lower than the surrounding tree tops. The time resolution was 5 min. The APS data are available from May 2004 onwards.

Meteorological measurements and back-trajectory calculations

Meteorological parameters (temperature, wind speed, wind direction, relative humidity, precipitation, rain intensity, atmospheric pressure, and global radiation) are measured continuously at the SMEAR II site. The half-hourly data were averaged into daily values and these were used in further analyses. To evaluate the effect of the ecosystem activity on PBAP concentrations, a thermal "growing season" was calculated (Lahti and Rönkä 2006). The growing season was defined as a continuous stretch of days with 24-h average temperatures above 5 °C. Here we also define a "pollen season", as a period when ecosystems produce pollen or other particles of biological origin locally. A "PBAP season" is the whole period when either locally-produced or long-range-transported pollen or other particles of biological origin are observed (Jato *et al*. 2006).

Long-range transport of the pollen and other particles of biological origin was studied with air mass history using back-trajectories. The 96-h back-trajectories were calculated with the HYSPLIT 4.8 model (Draxler and Hess 2004). This model was run with the GDAS meteoro-

logical archive produced by the US National Weather Service's National Centre for Environmental Prediction (NCEP) and archived by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (https://ready.arl.noaa.gov/archives.php).

Data analysis

The data from the first observation of PBAP each year until the end of the PBAP season were included into the analysis. The duration of the PBAP season was estimated separately for the pollen grains and the other particles of biological origin (Table 1). We classified the whole dataset into number concentration bins and analysed their frequency distributions over the season. There were numerous incidences with zero pollen counts as towards the end of the season pollen in the air is often scarce (Appendix 5). The number concentration distributions of other PBAP were clearly bimodal, and the frequency peaks of the concentration mode are shifted towards greater concentrations (Appendix 5). The relationship between PBAP concentration and particle mass was analyzed using the daily PBAP concentrations and the cumulative 2–3 day values for the corresponding PM_{10} mass concentration (according to the impactor's time resolution), and only the days when PBAP were recorded were included in the analysis. In order to evaluate the effect of meteorological parameters on PBAP number concentrations, the whole PBAP season, with days of zero concentrations, was included in the analysis. The relationship was analyzed using Pearson's correlation.

Results and discussion

Long-range transport and first observation of PBAP

In both studied years (2003 and 2004), the PBAP season started in March (Table 1). The first pollen concentrations, however, were recorded already weeks before the growing season started. Back trajectory analysis suggested that air masses arriving in Hyytiälä at that time originated from

regions where the growing season had already begun. The air masses originating from central Europe and travelling in close proximity to the surface layer during transport were seen to carry biological particles to Hyytiälä. The first recorded pollen of the 2004 season [pollen peaks just left of the dashed line in Fig. 2b; e.g. *Alnus* spp. (alder), *Populus* spp. (aspen), *Pinus* spp. (pine) and *Betula* spp. (birch) pollen] coincided with the air mass arrival from the south. In contrast, pollen was not observed in Hyytiälä when the air mass originated from the east (central Siberia), where the growing season had not yet begun, although altitude profiles of arriving air showed contact with the surface en route. The pollen and spores can be transported over long distances due to their ability to remain aerodynamically buoyant,

forestalling their deposition either due to gravity, wash out by rain, or impaction on terrestrial surfaces. Bioaerosols have also been observed to be transported several kilometres vertically up in the troposphere, where they can influence the cloud formation (DeLeon-Rodriguez *et al.* 2013). The changing climate may enhance convection, which might lead to more significant long-range transport of the biological particles in the future.

For the onset of pollen season, i.e. local pollen dispersal in wind-pollinating species, favourable meteorological conditions are necessary. Later spring in 2003, as compared with 2004, is likely linked with the later appearance of large numbers of pollen in 2003 (Fig. 2 and Table 1). Based on our results, we can conclude that the concentrations of other particles of bio-

Table 1. Characterization of wind-dispersed-PBAP (primary biological aerosol particles) seasons in 2003–2004 with respect to meteorological conditions and season duration. Thermal growing season is defined as 5 consecutive days with the mean temperature above 5 °C, pollen season as the period when ecosystems produce pollen or spores locally, and the PBAP season as the period when continuous elevated concentrations of wind-dispersed PBAP were observed. Note that values presented for other PBAP in 2004 do not represent the full PBAP season[a].

	Pollen grains		Other PBAP	
	2003	2004	2003	2004[a]
Season durations				
Growing season	4 May–2 Oct.	15 Apr.–8 Oct.	4 May–2 Oct.	15 Apr.–8 Oct.
Pollen season	23 Mar.–26 Aug.	10 Mar.–28 Sep.	9 Mar.–1 Dec.	3 Mar.–[a]
Length of pollen/PBAP season (days)	151/156	176/200	151/267	176/>154
PBAP peaks				
Day of max. concentration	13 May	8 May	23 Aug	1 Jul
Max. daily number concentration (m^{-3})	9741	2013	145612	72074
Max. daily mass concentration (μm m^{-3})[b]	98	26	2.5	1.2
Cumulative concentrations				
Number concentration (Σ m^{-3})	40870	18025	3713792	> 2572449
Mass concentration (Σ μm m^{-3})[b]	898	484	63	> 44
Temperature (°C)[c]				
Mean	10.9	9.9	8.9	9.0
Mean daily min.	7.1	6.0	4.8	4.9
Mean daily max.	14.6	13.4	11.2	12.7
Humidity (%)[c]	70.3	64.1	75.7	62.6
Solar radiation at surface (W m^{-2})[c]	93	69	46	91
Precipitation (mm day^{-1})[c]	1.7	1.9	1.7	2.0

[a] Pollen was examined for 7 Mar.–29 Aug. 2003 and 3 Mar.–30 Sep. 2004 including the complete PBAP season for both years, whereas the other PBAP were analysed for 6 Mar.–31 Dec. 2003 and 3 Mar.–4 Aug. 2004.

[b] Mean daily pollen and fungal spore mass concentrations (*m*) were calculated using the following equation: $m = N \times 4/3\pi \times (D/2)^3 \times \sigma$, where *N* is the pollen or spore number concentration (m^{-3}), *D* (μm) is either the pollen diameter (values taken from Appendix 1), or the spherical spore diameter, assumed to be 3 μm for all species (corresponding to typical aerodynamic diameter for most common spores; *see* Reponen *et al.* 2001, Hussein *et al.* 2013, Schumacher *et al.* 2013), and σ is the pollen or spore density here assumed to be 1.4 g cm^{-3} or 1.2 g cm^{-3}, respectively (*see* Reponen 1995, Jacobson and Streets 2009).

[c] Mean for the whole PBAP season.
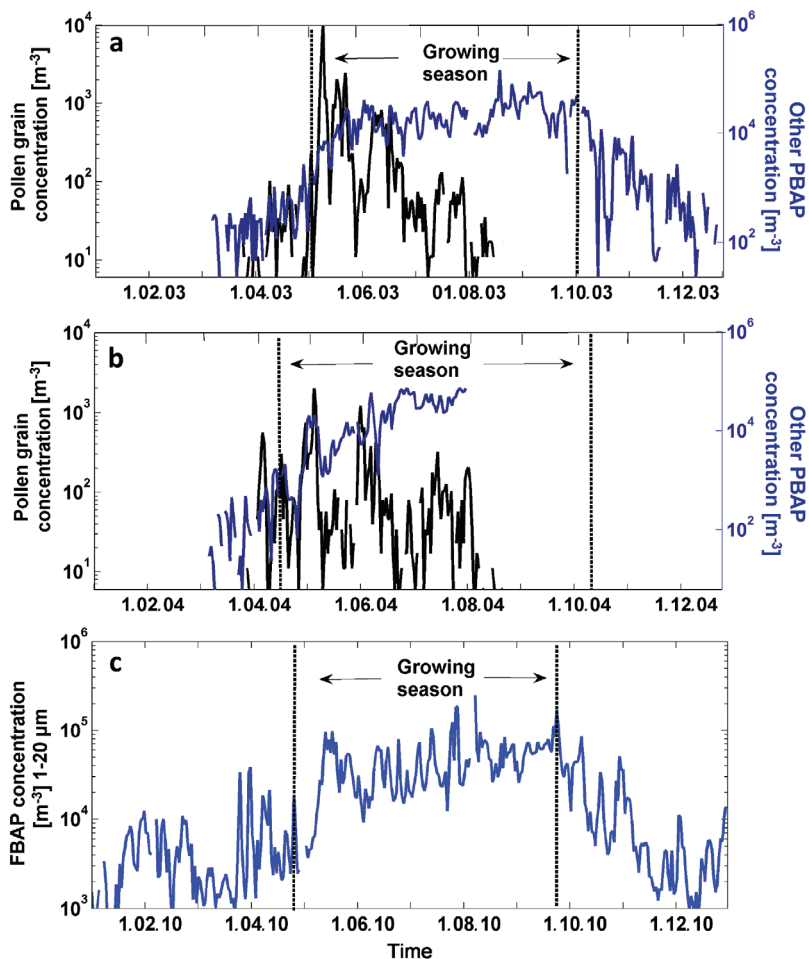
Fig. 2. Concentrations of airborne pollen and other primary biological aerosol particle (PBAP) (daily sums) in Hyytiälä in (**a**) 2003 and (**b**) 2004. The dashed lines indicate the beginning and the end of the growing season for each year. (**c**) Concentrations of airborne fluorescent biological aerosol particles (FBAP, daily means integrated over 1–20 µm) in Hyytiälä in 2010 [data from Schumacher *et al.* (2013)].

logical origin are more consistent throughout the year, indicating that the spore production by fungi is less closely related to spring advancement and preceding, as well as to prevailing temperatures. This also indicates that long-range transport of other PBAP is not as evident. The long-range transport does not take place only at the beginning of the PBAP season, but also during the local pollen season. Thus, long-distance transport will always add to the local pollen and spore number concentrations.

To evaluate the importance of local sources and the effect of the land use close to Hyytiälä, we studied the PBAP concentrations as a function of local wind direction. The total concentrations showed no dependence on the local wind direction, however for individual species, wind-dependent differences in concentration were found (Fig. 3). Especially *Urtica* spp. (nettles) and *Artemisia* spp. (mugwort) pollen counts were higher when local wind arrived from the south or southeast, whereas *Picea* spp. (spruce) and *Betula* spp. pollen was recorded mainly from the south and southwest (Fig. 3b). This is in line with the land cover map (Fig. 1) which illustrates that the southeast sector has more scrub and herbaceous vegetation and agriculture (e.g. natural fields with nettles and mugwort), whereas the southwest sector has a higher percentage of broad-leaved and mixed forest (e.g. birch), and the coniferous forest is prevailing in the northwest direction (Williams *et al.* 2011). Not surprisingly, this observation confirms that the particles measured at a given site are strongly influenced both by the flora in the surroundings and by the local meteorology.
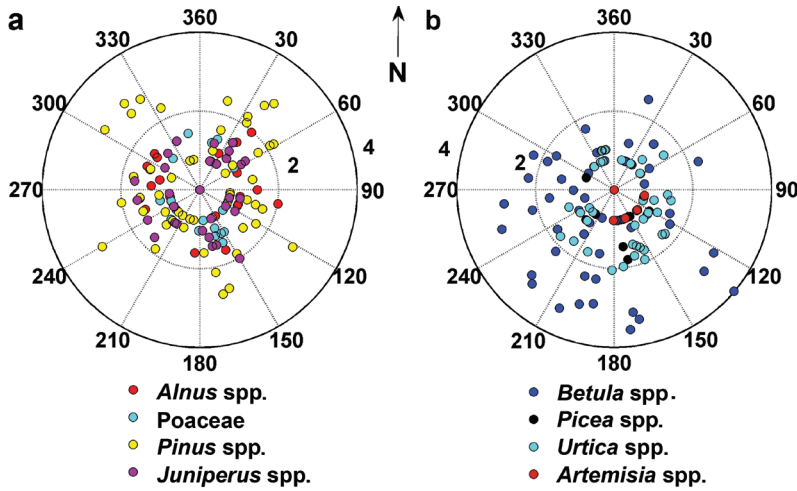
Fig. 3. Pollen concentration [log(grains per m³)] as a function of local wind direction in 2003. (**a**) Species whose pollen concentrations are independent of local wind direction, and (**b**) species, whose pollen originates mostly from wind directions 120°–270°.
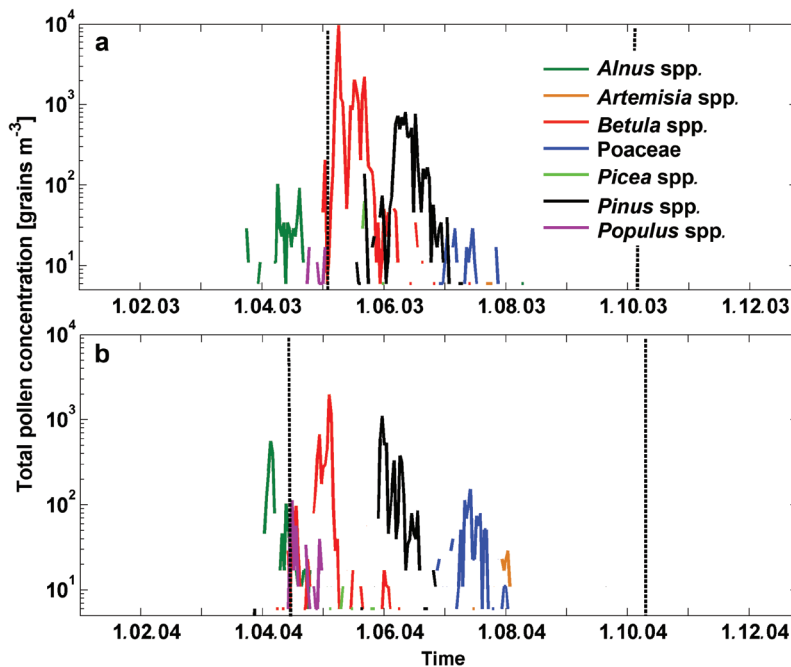
Fig. 4. Pollen concentrations of dominant taxa in Hyytiälä in (**a**) 2003 and (**b**) 2004. The dashed lines show the beginning and the end of the growing season.

Patterns in pollen concentrations

The highest peaks in airborne pollen concentrations were measured in both years in May and June (*see* Appendices 1–2). The highest daily peak concentrations of *Betula* spp. and *Pinus* spp. pollen exceeded 9700 and 800 grains per m³, respectively (Table 2). The seasonal patterns in pollen concentrations in 2003–2004 for the dominant taxa follow a clear cycle (Fig. 4), as has been shown in many previous studies (e.g.

Siljamo *et al.* 2008, Yli-Panula *et al.* 2009). In winter, the air is free of pollen in boreal latitudes. Most tree species flower during spring, whereas most herbaceous species do so later in the summer. The timing and the sequence of flowering of the different species takes place in same order each year, regardless of environmental factors. The pollen concentration patterns in the study years were very similar, but their absolute concentrations may also differ substantially from year to year, depending on meteorological

conditions in the previous year. In case of a substantial flowering and pollen production and thus an increased seed production in previous year, the catkin production is often decreased due to limited resources in the next year. The onset of growing season affects greatly pollen number concentrations: in 2003 the majority of first pollen was observed considerably later than in 2004, due to the later onset of the growing season by three weeks. However, the difference between the dates when the maximum pollen numbers were registered in both years was only five days, which indicates that in the case of late spring the flowering was commencing much faster than in an early spring, due to the rapid increase in daily mean temperatures (Table 3).

The most common sources for the pollen in boreal forest were *Betula* spp., *Pinus* spp., *Urtica* spp. and *Alnus* spp. Our observations are consistent with a report by Rantio-Lehtimäki *et al*. (1994) who showed peak concentrations of pollen grains from *Betula* spp. sampled in Turku, Finland, of > 10^3 m^{-3} for most of May 1985, peaking at ~10^4 m^{-3} in mid-month. We measured a large fraction of the total pollen count of the year during a single day. In 2004, the peak values of 31% and 23% of the season's total birch and pine pollen counts, respectively, were measured on one day, whereas in 2003 the highest concentration on a single day was 10% for both species (Table 2). Most plants tend to open their flowers on the first favourable day, usually

Table 2. Characteristics of pollen in 2003 and 2004 in Hyytiälä.

	First observation	Last observation	Maximum number recorded on	Maximum number (grains m^{-3})	Season duration (days)	Cumulative sum (grains m^{-3}) and percentage
2003						
Alnus (alder)	23 Mar	4 May	12 Apr	103	42	659 (2%)
Artemisia (mugwort)	21 Jul	30 Jul	28 Jul	6	9	30 (< 1%)
Betula (birch)	31 Mar	2 Aug	13 May	9741	124	29763 (72%)
Chenopodium (goosefoot)	–	–	–	–	–	–
Poaceae (grasses)	3 Jul	26 Aug	11 Jul	29	54	280 (< 1%)
Juniperus (juniper)	26 May	7 Jul	9 Jun	103	92	606 (1%)
Picea (spruce)	25 May	25 Jun	26 May	63	31	144 (< 1%)
Pinus (pine)	31 Mar	26 Aug	16 Jun	807	48	7986 (20%)
Populus (aspen)	27 Apr	10 May	28 Apr	17	13	81 (< 1%)
Quercus (oak)	9 Jun	9 Jun	11 Jun	6	1	6 (< 1%)
Rumex (sorrel)	24 Jun	17 Aug	8 Jul	11	54	41 (< 1%)
Salix (willow)	8 May	4 Jun	10 May	11	27	59 (< 1%)
Ulmus (elm)	–	–	–	–	–	–
Urtica (nettles)	2 Jul	26 Aug	21 Jul	109	55	1068 (3%)
All species	23 Mar	26 Aug	13 May	9741	156	40870 (100%)
2004						
Alnus (alder)	5 Apr	9 May	10 Jun	561	34	1979 (11%)
Artemisia (mugwort)	18 Jul	6 Sep	7 Aug	29	50	156 (< 1%)
Betula (birch)	11 Apr	12 Aug	8 May	1979	123	6369 (35%)
Chenopodium (goosefoot)	5 Aug	5 Aug	5 Aug	6	1	6 (< 1%)
Poaceae (grasses)	22 Jun	25 Aug	19 Jul	154	64	1132 (6%)
Juniperus (juniper)	17 Apr	21 Jul	4 Jun	103	95	423 (2%)
Picea (spruce)	8 May	7 Jun	15 May	11	30	83 (< 1%)
Pinus (pine)	16 Mar	26 Sep	4 Jun	1104	194	4779 (27%)
Populus (aspen)	10 Mar	6 May	19 Apr	114	57	399 (2%)
Quercus (oak)	4 Jun	4 Jun	4 Jun	6	1	6 (< 1%)
Rumex (sorrel)	13 Jun	7 Aug	18 Jun	11	55	93 (< 1%)
Salix (willow)	18 Apr	25 Jun	20 May	86	68	891 (5%)
Ulmus (elm)	2 May	2 May	2 May	6	1	6 (< 1%)
Urtica (nettles)	13 Jun	26 Aug	6 Aug	166	74	1488 (8%)
All species	10 Mar	26 Sep	8 May	2013	200	18025 (100%)

when there is no rain, temperature is high, sunshine is moderate and wind speed is low (Jones and Harrison 2004). In boreal forest, especially, *Picea* spp., *Populus* spp. and *Salix* spp. show this behaviour as the peak pollen emissions are seen in the beginning of their pollen season (Table 3).

In comparison with the main pollen-producing species, e.g. *Quercus* spp. and *Ulmus* spp. pollen concentrations are almost all the time close to the lower detection limit of the Burkard trap. We already know that very few elms and oaks grow in the region so their pollen is most likely transported with air masses to Hyytiälä. Similarly, the *Chenopodium* spp. pollen was poorly represented in the samples indicating that the goosefoot pollen is only seldom lifted up to the sample level above the canopy.
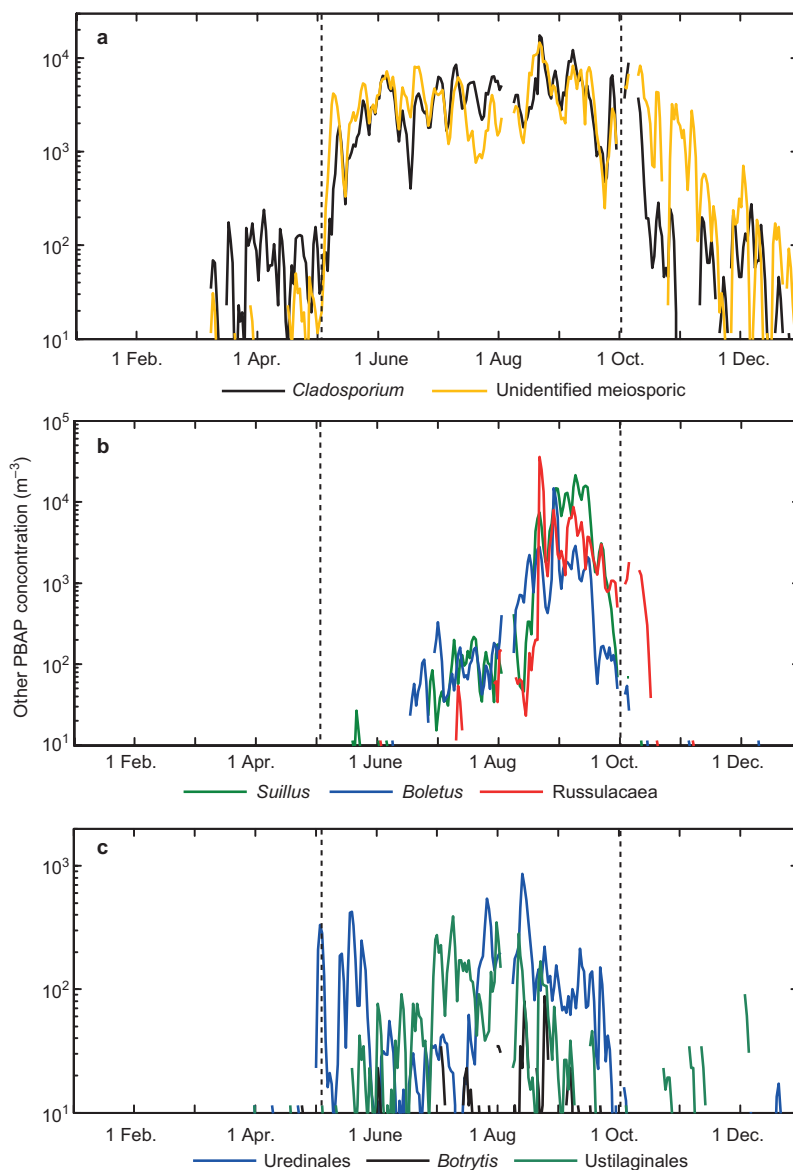
Patterns in other PBAP concentrations

The total number concentrations of other PBAP such as fungal spores are much higher as compared with those of the pollen (10^4–10^6 m^{-3} and 10^2–10^4 m^{-3}, respectively; Fig. 2). It should be noted that the measured concentrations of both pollen grains and other particles of biological origin are well above average concentrations in vegetated regions reported by Després *et al.* (2012). In contrast to the pattern in pollen counts, the peaks in other PBAP concentrations were detected in August and September, which is in agreement with the measurements of fluorescent aerosol carried out by Schumacher

et al. (2013) in 2010 at the same site (Fig. 2). The distribution of other PBAP was very similar in both years. The highest concentrations were measured between 1 May and 1 November 2003, whereas in winter season the concentrations were lower (*see* Appendices 3–4). The daily spore concentrations of the most common taxa, Russulaceae and *Boletus* spp. exceeded 7.1 $\times 10^4$ and 2.8 $\times 10^4$ spores m^{-3}, respectively, on the day with the highest peak. The particles of biological origin (e.g., fungal, algal, and bacterial spores) are a ubiquitous component of the atmosphere, though daily concentrations vary by $> 10^2$ from winter minimum to an autumn maximum (Fig. 2) exceeding 10^5 m^{-3}. Schumacher *et al.* (2013) suggested that the daily variability of FBAP at the Hyytiälä site was consistent with the trend expected for fungal spore emissions, and several other studies have suggested the relative dominance of fungal spore concentrations among biological particle classes in clean, vegetated environments (e.g. Gabey *et al.* 2011, Huffman *et al.* 2012). Our results confirm this.

Most common fungal spores identified were those of *Cladosporium*, Basidiomycota and Ascomycota. Many common fungi in Ascomycota and Basidiomycota actively eject their spores with liquid jets or droplets (osmotic pressure and surface tension effects), whereas others rely on dry spore detachment by wind or other external forces. Similarly as for the pollen species, the duration of the total airborne period differed among the spore species (Fig. 5). For example, spore concentrations of *Cladosporium*

Table 3. The total particle mass (PM$_{total}$, μg m^{-3}) and the particle mass in different size fractions (PM$_1$, PM$_{1–2.5}$, PM$_{2.5–10}$ and PM$_{>10}$; μg m^{-3}), measured in four seasons with PM10-impactor in Hyytiälä 2003–2004. Winter = December–February, spring = March–May, summer = June–August, autumn = September–November.

|	PM$_1$	PM$_{1–2.5}$	PM$_{2.5–10}$	PM$_{>10}$	PM$_{total}$
2003					
Winter	4.099	1.965	0.875	0.198	7.137
Spring	6.357	1.874	1.209	0.633	10.074
Summer	6.490	1.467	1.408	1.707	11.072
Autumn	2.718	1.334	1.324	0.368	5.744
2004					
Winter	3.235	0.990	1.043	0.103	5.371
Spring	3.512	0.900	0.884	0.746	6.042
Summer	3.549	1.105	1.217	1.010	6.881
Autumn	3.357	1.160	1.428	0.467	6.412

Fig. 5. Concentrations of other primary biological aerosol particles (PBAP) of different taxa in 2003 in Hyytiälä. Three patterns are seen: (**a**) spores which occur around the year, high concentrations prevail during the whole growing season (*Cladosporium* spp, unidentified meiosporic), (**b**) spores whose peak concentrations are closer to the end of the growing season (*Suillus* spp, *Boletus* spp, Russulaceae), and (**c**) spores which are observed almost solely during the growing season (*Botrytis*, Uredinales, Ustilaginales). The dashed lines show the start and the end of the growing season.

spp. were above zero on consecutive days between 26 April and 10 October 2003 (Fig. 5a), while those of *Suillus* spp. only between 4 August and 30 September 2003 (Fig. 5b). This is naturally a consequence of the phenological timing of the *Basidiomycota* fruiting bodies later in the summer, whereas the *Cladosporium* (including e.g. many common moulds) are present all the time in dead plant litter, soil and on various surfaces.

Contribution of pollen and other PBAP to particle mass

The total suspended particulate matter (PM) mass exhibited a clear pattern (Table 3 and Fig. 6). The PM mass was found to peak during the spring (March–May) and summer (June–August); the highest average mass concentration measured during the studied period was during the summer of 2003 when ultrafine PM_1 peaked at 6.5 μg m^{-3} and $PM_{>10}$ at 1.7 μg m^{-3}. In contrast, during the
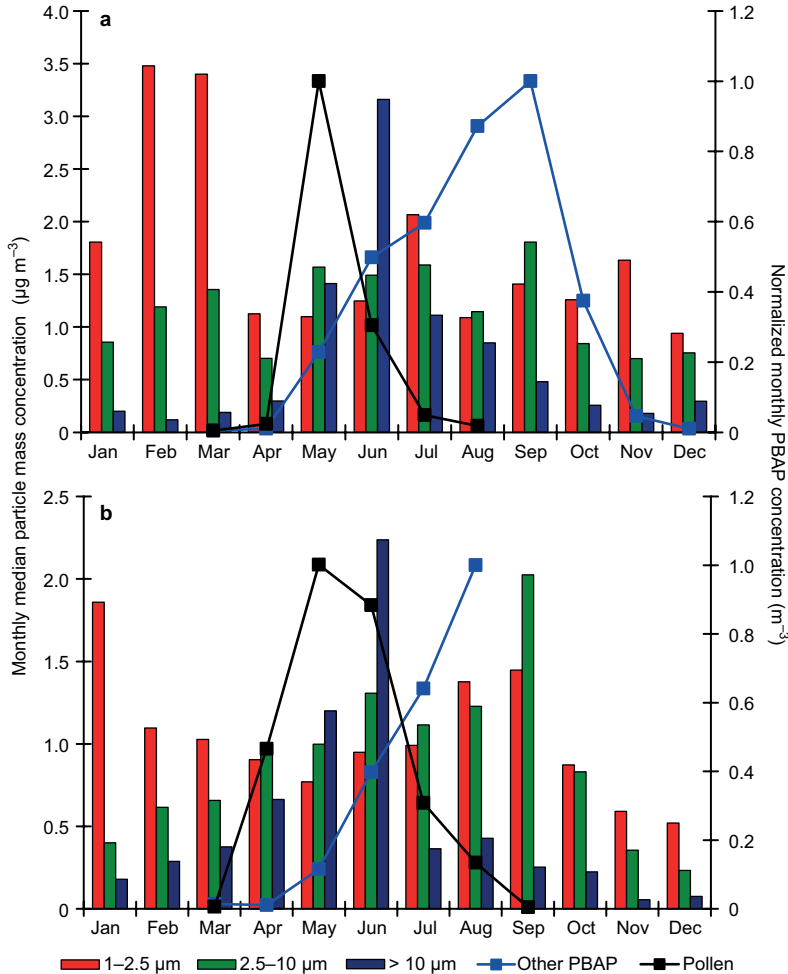
Fig. 6. Monthly median particle mass concentration of different size fractions, and normalized monthly total pollen and other PBAP concentrations measured in Hyytiälä in (**a**) 2003 and (**b**) 2004.

summer 2004 the maximum mass was measured in the PM$_{>10}$ fraction (1.0 μg m^{-3}), whereas the PM$_{2.5-10}$ fraction (1.4 μg m^{-3}) was dominant during the autumn (September–November). Thus, a common feature for both years was the peak concentration of PM$_{>10}$ during summer and the peak PM$_{2.5-10}$ during autumn.

Our results support those of Laakso *et al.* (2003) who measured particle mass in Hyytiälä between 1999 and 2001, and found spring and autumn maxima in particle mass in the size range of sub-10 μm and in particle number in the accumulation mode (0.1–1 μm). However, they suggested that the peaks in particle mass could be non-biological, and explained by the long-range-transported PM, resulting from street sanding, and the use of studded tires. They did not report the PM for different size fractions but

only the total of PM$_1$, PM$_{2.5}$ and PM$_{10}$, and thus, the emissions of PBAP were not considered as a potential explanation for the high PM concentrations during the pollen season. Based on our results, the pattern in the particle mass from natural biological sources is clearly observed at the Hyytiälä site, as Hyytiälä represents a rural background site with a mean annual PM$_{10}$ concentration of 6.9 μg m^{-3} (Laakso *et al.* 2003). Furthermore, during the peak pollen season, the impactor plates were visibly yellow due to the pollen particles, indicating their large contribution to particle mass.

The size of a typical pollen grain is in the range 10–100 μm (Appendix 1), whereas other particles of biological origin are much smaller, 1–50 μm (mostly 2–10 μm; e.g. Hussein *et al.* 2013; Appendix 3). While looking at the

monthly values, a peak in the concentrations of other particles of biological origin was detected at the same time as the autumn maxima in $PM_{2.5-10}$, whereas the peak in the pollen concentration coincided with the $PM_{>10}$ peak in summer (Fig. 6). In the boreal forest, the highest PM values were measured in June (Fig. 6), when both the pollen and spores contribute to the particle mass. As soon as airborne pollen concentrations begun to decline and the pollen season ended, the large PM fractions started to decrease as well, indicating that in a rural environment where background mass concentration is low, primary biological aerosol can have a major contribution to the total particle mass.

The other particles of biological origin dominated the PBAP particle number in Hyytiälä, but the pollen was more pronounced in particle mass. Using cumulative number concentrations (*see* Table 1), the contribution of the other particles of biological origin was ~99% and that of pollen ~1% of the total PBAP number. On the other hand, of the cumulative total PBAP mass concentrations, pollen represents ~93% and other particles of biological origin ~7% (Table 1). Assuming species-specific pollen grain sizes according to Pöhlker *et al.* 2013 and www.polleninfo.org, an average spore size of 3.0 μm for the other PBAP (e.g. Jacobson and Streets 2009, Hussein *et al.* 2013, Schumacher *et al.* 2013), and a spherical shape for pollen and spore particles with a density of 1.4 and 1.2 g cm^{-3} (Reponen *et al.* 1995, Jacobson and Streets 2009), respectively, one can calculate an order-of-magnitude estimation of the cumulative particle mass contribution from PBAP in 2003. This shows a daily mean of 6.3 μg m^{-3} mass of these PBAP classes during the pollen season (5.9 and 0.4 μg m^{-3} for pollen and other PBAP, respectively). This corresponds to an average mass fraction of 65% with respect to the total PM mass (again 60% and 5% for pollen and other PBAP, respectively). Previous works have shown that number and mass concentrations of fungal spores are typically ~10^3–10^4 m^{-3} and ~0.1–1 μg m^{-3}, respectively, in the continental boundary layer (Elbert *et al.* 2007, Fröhlich-Nowoisky *et al.* 2009), whereas the pollen grain number and mass concentrations are ~10–10^3 m^{-3} and ~1 μg m^{-3}, respectively (Sofiev *et al.* 2006,

Fröhlich-Nowoisky *et al.* 2009). These values correspond well with our results.

The relationship between PM and PBAP number concentrations was also analysed using 2–3 day averages (Fig. 7). The pollen concentrations correlated moderately with $PM_{2.5-10}$ ($r_P = 0.53$, $p < 0.05$, $n = 136$) and $PM_{>10}$ ($r_P = 0.63$, $p < 0.05$, $n = 136$). The corresponding coefficient values for PBAP concentrations are $r_P = 0.52$ ($p < 0.05$, $n = 277$) and $r_P = 0.45$ ($p < 0.05$, $n = 277$), respectively. This is in line with typical pollen and spore particle sizes (e.g. Hussein *et al.* 2013). The relationship between the PBAP number concentrations measured with the spore trap and the total particle number measured with the APS was not as clear. The APS measures size distribution of 0.5–20 μm particles which represents the coarse mode (> 1 μm) and upper end of the accumulation mode (0.1–1 μm), yielding the total number concentration but no information about particle origin. Only the smallest intact bioaerosols such as viruses and bacteria would likely be visible in accumulation mode size range, however cell fragments, for example pollen grains that are ruptured after uptake of water, or one-cell Ascomycota could contribute large numbers to the accumulation mode during pollen season and during periods of high humidity (e.g. Taylor *et al.* 2004, Miguel *et al.* 2006). Based on our observations regarding particles > 1 μm, we suggest that pollen and other PBAP both contribute significantly to the total particle mass during the pollen season, but that their influence on the total particle number in Hyytiälä is only minor.

Effect of local meteorology

Meteorological factors affect both the initial release of the material and its dispersal once airborne, and thus affect the atmospheric bioaerosol concentrations (Jones and Harrison 2004). We studied the effect of local meteorological variables on the PBAP concentrations in boreal forest by means of Pearson's correlation between PBAP and meteorological parameters (temperature, global radiation, relative humidity, rain intensity and wind speed). In these analyses, the daily mean values were used, which is a rather rough method
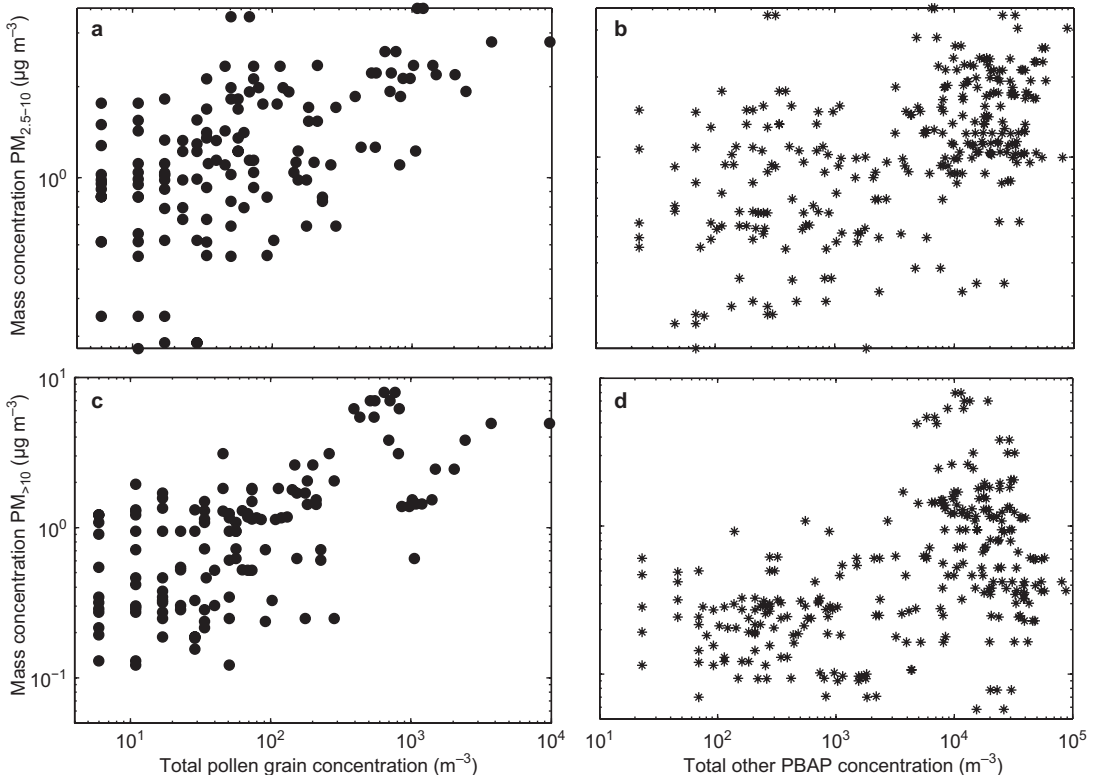
Fig. 7. Relationships between 2–3 day average particle mass concentrations of $PM_{2.5-10}$ (**a**, **b**) and $PM_{>10}$ (**c**, **d**), and total pollen (**a**, **c**) or other PBAP number concentrations (**b**, **d**) during the PBAP season in Hyytiälä.

of testing the relation between meteorological factors and fungal spore concentrations. A moderate correlation was found between other PBAP and mean daily temperature ($r_p = 0.60$, $p < 0.05$, $n = 423$), which indicates that higher spore concentrations are associated with higher air temperatures (Fig. 8b). A weak relationship was also found with rain intensity ($r_p = 0.28$, $p < 0.05$, $n = 423$) and wind speed ($r_p = -0.22$, $p < 0.05$, $n = 423$) at the canopy level (18 m).

Most of the spore plumes were detected on days without rain, but also during intense rain episodes, increased number concentrations were measured. Recent findings from a semi-arid forest in North America indicate that rainfall can trigger intense bursts of bioaerosol emission and massive enhancements of atmospheric bioaerosol concentrations by an order of magnitude or more (Huffman *et al.* 2013, Prenni *et al.* 2013). Further, Schumacher *et al.* (2013) showed that the fluorescent bioaerosol concentrations in Hyytiälä increased during summer rain events.

Although in our study no correlation between PBAP concentrations and relative humidity (RH) was found, high spore concentrations were sometimes associated with high RH values. In summer the spore concentrations were increasing with RH, but at very high RH (> 85%) the concentrations started to decrease. Thus, temperature and RH seem to affect concentration of particles of biological origin, such as spores (e.g. Elbert *et al.* 2007).

The daily mean pollen grain numbers showed no clear dependence on any of the studied meteorological variables (Fig. 8a). We recorded the highest pollen concentrations on relatively warm days, with moderate RH, no rain, both under clear and cloudy skies. Lo and Levetin (2007) found positive correlations between pollen and daily temperature and negative correlations between pollen and precipitation. Ogden *et al.* (1969) showed that pollen emission was reduced in the presence of rain or when the wind speed was low. The pollen is often ejected in clumps
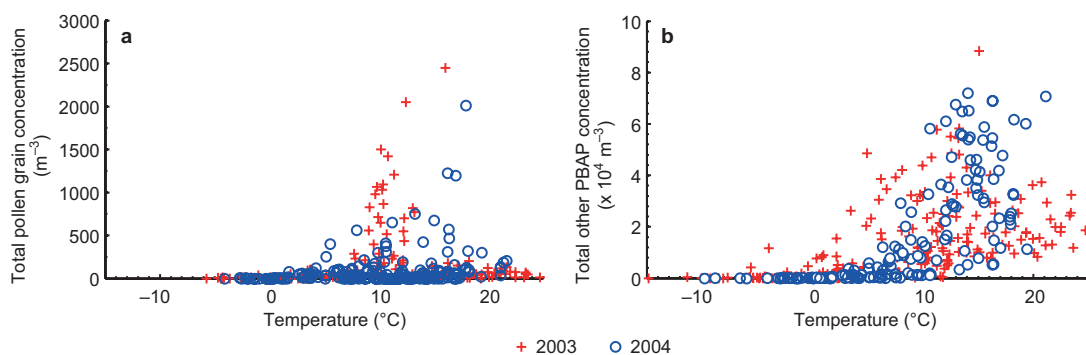
Fig. 8. Relations between (**a**) total pollen grain concentration and (**b**) other PBAP concentration, and daily mean temperature in 2003–2004 in Hyytiälä during the PBAP season.

that stick to their neighbouring vegetation and are blown away after drying (Jones and Harrison 2004). The concentrations of other PBAP changed considerably from day to day, which might indicate that changes in the local meteorological conditions could have a larger effect on wind-dispersed spores compared to pollen (Figs. 4 and 5).

It should be noted that different meteorological factors may be important for different particles of biological origin or pollen taxa. We noticed that the temperature dependence was more pronounced for some taxa than for the others (data not shown). Also, most probably the time resolution of the data (daily averages) was not sufficient enough to properly reveal the short-term effects of meteorological variables on emissions of other PBAP. Parameterization of biogenic aerosols in various environments would be important for estimating the global and regional effects of PBAPs on climate and hydrological cycle.

Conclusions

We showed that the presence of wind-dispersed pollen follows a clear seasonal pattern according to the pollen seasons of the source plants, but that the other PBAP are more evenly found throughout the year. The observed particle mass concentrations exhibited a cycle, with $PM_{2.5-10}$ peaking in autumn and $PM_{>10}$ peaking in summer. This cycle in coarse particle mass exhibited similar temporal behaviour as occurrence of PBAP in the boreal forest, suggesting that bioaerosols

contribute significantly to the total aerosol mass burden in such environments. Only the number concentrations of PBAP were measured here, and not particle size or morphology, thus limiting an accurate estimation of PBAP mass by this method. However, numbers reported here are in good agreement with the values available from literature, although detailed numbers from the boreal region had not been reported previously.

Recently, PBAP have been studied in great detail due to their enhanced capability to act as ice nuclei (e.g. Morris *et al*. 2004, Möhler *et al*. 2007, DeMott *et al*. 2011). As a result, there is currently much discussion about the potential of biological particles to contribute to atmospheric processes of the formation and evolution of ice clouds and precipitation. For example, studies have shown that the Amazon rainforest in Brazil provides a pristine source of natural cloud nuclei that feeds its own hydrological cycle (Pöschl *et al*. 2010, Morris *et al*. 2014). The high fraction of PBAP in the coarse mode presented here suggests that PBAP is an important component of natural aerosol also in the boreal region that should be considered when modelling cloud and precipitation effects.

This study was focused on both pollen and spore concentrations, providing long-term field measurement data of biological aerosol concentrations on seasonal scales. Such data can be used to include the bioaerosol concentrations in the global aerosol models. However, it should be noted that sampling of pollen, other particles of biological origin, and other bioaerosols lacks standardized sampling and analysis procedures,

which may lead to vast uncertainties and very different recorded bioaerosol concentrations. Here, the method for biological aerosol particle sampling followed the EAN standard. While estimating the influence of PBAP on the total particle load, the daily cumulative concentration and length of seasonal period of high pollen and spore concentration are probably more relevant than daily mean or monthly mean values.

An important task for the future research is to connect PBAP to other ecosystem processes e.g. formation of biogenic volatile organic compounds, and to study the feedback mechanisms linking forest, aerosols, and climate (Kulmala *et al.* 2004, Lappalainen *et al.* 2009). Hakola *et al.* (2006) showed that the maximum volatile organic compound emissions from pine shoots occurred concomitant with the high concentration of airborne pathogen spores, suggesting a potential defensive role of the sesquiterpene emissions. These need to be investigated in follow-up studies.

References

Barnes C., Schreiber K., Pacheco F., Landuyt J., Hu F. & Portnoy J. 2000. Comparison of outdoor allergenic particles and allergen levels. *Ann. Allergy Asthma Immunol.* 84: 47–54.

Bassett I.J, Barnes C.W. & Parmelee J.A. 1978. *An atlas of airborne pollen grains and common fungus spores of Canada*. Monograph no. 18, Research Branch, Canada Department of Agriculture.

Battarbee J.L., Rose N.L. & Long X. 1997. A continuous, high resolution record of urban airborne particulates suitable for retrospective microscopical analysis. *Atmos. Environ.* 31: 171–181.

Beggs P.J. 2004. Impacts of climate change on aeroallergens: past and future. *Clin. Exp. Allergy.* 34: 1507–1513.

Burge H. 1990. Bioaerosol — prevalence and health effects in the indoor environment. *J. Allergy Clin. Immunol.* 86: 687–701.

Deguillaume L., Leriche M., Amato P., Ariya P.A., Delort A.-M., Pöschl U., Chaumerliac N., Bauer H., Flossmann A.I. & Morris C.E. 2008. Microbiology and atmospheric processes: chemical interactions of primary biological aerosols. *Biogeosciences* 5: 1073–1084.

DeLeon-Rodriguez N., Lathem T.L., Rodriguez-R L.M., Barazesh J.M., Anderson B.E., Beyersdorf A.J., Ziemba L.D., Bergin M., Nenes A. & Konstantinidis K.T. 2013. Microbiome of the upper troposphere: Species composition and prevalence, effects of tropical storms, and atmospheric implications. *Proc. Natl. Acad. Sci. USA* 110: 2575–2580.

DeMott P.J., Möhler O., Stetzer O., Vali G., Levin Z., Petters M.D., Murakami M., Leisner T., Bundke U., Klein H., Kanji Z.A., Cotton R., Jones H., Benz S., Brinkmann M., Rzesanke D., Saathoff H., Nicolet M., Saito A., Nillius B., Bingemer H., Abbatt J., Ardon K., Ganor E., Georgakopoulos D.G. & Saunders C. 2011. Resurgence in ice nuclei measurement research. *Bull. Am. Meteorol. Soc.* 92: 1623–1635.

Després V.R., Huffman J.A., Burrows S.M., Hoose C., Safatov A.S., Buryak G.A., Fröhlich-Nowoisky J., Elbert W., Andreae M.O., Pöschl U. & Jaenicke R. 2012. Primary biological aerosol particles in the atmosphere: a review. *Tellus* 64B, 15598, doi:10.3402/tellusb.v64i0.15598

Diehl K., Quick C., Matthias-Maser S., Mitra S.K. & Jaenicke R. 2011. The ice nucleating ability of pollen — Part I: laboratory studies in deposition and condensation freezing modes. *Atmos. Res.* 58: 75–87.

Draxler R.R. & Hess G.D. 2004. *Description of the HYSPLIT 4 modelling system*. NOAA Technical Memorandum ERL ARL-224.

EEA 1997. *Technical and methodological guide for updating CORINE land cover data base*. European Environment Agency, EUR 17288 EN.

Elbert W., Taylor P.E., Andreae M.O. & Pöschl U. 2007. Contribution of fungi to primary biogenic aerosols in the atmosphere: wet and dry discharged spores, carbohydrates, and inorganic ions. *Atmos. Chem. Phys.* 7: 4569–4588.

Fröhlich-Nowoisky J., Pickersgill D.A., Després V.R. & Pöschl U. 2009. High diversity of fungi in air particulate matter. *Proc. Natl. Acad. Sci. USA* 106: 12814–12819.

Gabey A., Stanley W.R., Gallagher M. & Kaye P.H. 2011. The fluorescence properties of aerosol larger than 0.8 μm in urban and tropical rainforest locations. *Atmos. Chem. Phys.* 11: 5491–5504.

Gregory P.H. 1961. *The microbiology of the atmosphere*. Leonard Hill Books Ltd., London, New York.

Gregory P.H. 1978. Distribution of airborne pollen and spores and their long-distance transport. *Pure App. Geophys.* 116: 309–315.

Haapanala S., Rinne J., Hakola H., Hellén H., Laakso L., Lihavainen H., Janson R., O'Dowd C. & Kulmala M. 2007. Boundary layer concentrations and landscape

scale emissions of volatile organic compounds in early spring. *Atmos. Chem. Phys.* 7: 1869–1878.

Haga D.I., Iannone R., Wheeler M.J., Mason R., Polishchuk E.A., Fetch T.Jr., van der Kamp B.J., McKendry I.G. & Bertram A.K. 2013. Ice nucleation properties of rust and bunt fungal spores and their transport to high altitudes, where they can cause heterogeneous freezing. *J. Geophys. Res. Atmos.* 118: 7260–7272.

Hakola H., Tarvainen V., Bäck J., Ranta H., Bonn B., Rinne J. & Kulmala M. 2006. Seasonal variation of mono- and sesquiterpene emission rates of Scots pine. *Biogeosciences.* 3: 93–101.

Hari P. & Kulmala M. 2005. Station for Measuring Ecosystem–Atmosphere Relations (SMEAR II). *Boreal Env. Res.* 10: 315–322.

Heald C. & Spracklen D. 2009. Atmospheric budget of primary biological aerosol particles from fungal spores. *Geophys. Res. Lett.* 36: L09806, doi:10.1029/2009GL037493.

Hjelmroos M. 1991. Evidence of long-distance transport of *Betula* pollen. *Grana* 30: 215–228.

Hoose C., Kristjansson J.E. & Burrows S.M. 2010. How important is biological ice nucleation in clouds on a global scale? *Environ. Res. Lett.* 5: 024009, doi:10.1088/1748-9326/5/2/024009.

Huffman J.A., Treutlein B. & Pöschl U. 2010. Fluorescent biological aerosol particle concentrations and size distributions measured with an Ultraviolet Aerodynamic Particle Sizer (UV-17146 APS) in central Europe. *Atmos. Chem. Phys.* 10: 3215–3233.

Huffman J.A., Sinha B., Garland R.M., Snee-Pollmann A., Gunthe S.S., Artaxo P., Martin S.T., Andreae M.O. & Pöschl U. 2012. Biological aerosol particle concentrations and size distributions measured in pristine tropical rainforest air during AMAZE-08. *Atmos. Chem. Phys.* 12: 11997–12019.

Huffman J.A., Prenni A.J., DeMott P.J., Pöhlker C., Mason R.H., Robinson N.H., Fröhlich-Nowoisky J., Tobo Y., Després V.R., Garcia E., Gochis D.J., Harris E., Müller-Germann I., Ruzene C., Schmer B., Sinha B., Day D.A., Andreae M.O., Jimenez J.L., Gallagher M., Kreidenweis S.M., Bertram A. K. & Pöschl U. 2013: High concentrations of biological aerosol particles and ice nuclei during and after rain. *Atmos. Chem. Phys.* 13: 6151–6164.

Hirst J.M. 1952. An automatic volumetric spore trap. *Ann. Appl. Biol.* 39: 257–265.

Hussein T., Norros V., Hakala J., Petäjä T., Aalto P.P., Rannik U., Vesala T. & Ovaskainen O. 2013. Species traits and inertial deposition of fungal spores. *J. Aerosol. Sci.* 61: 81–98.

Ilvesniemi H., Levula J., Ojansuu R., Kolari P., Kulmala L., Pumpanen J., Launiainen S., Vesala T. & Nikinmaa E. 2009. Long-term measurements of the carbon balance of a boreal Scots pine dominated forest ecosystem. *Boreal Env. Res.* 14: 731–753.

Jacobson M.Z. & Streets D.G. 2009. Influence of future anthropogenic emissions on climate, natural emissions, and air quality. *J. Geophys. Res. Atmos.* 114: D08118, doi:10.1029/2008JD011476.

Jaenicke R. 2005. Abundance of cellular material and proteins in the atmosphere. *Science* 308: 73.

Jaenicke R., Matthias-Maser S. & Gruber S. 2007. Omnipresence of biological material in the atmosphere. *Environ. Chem.* 4: 217–220.

Jato V., Rodríguez Rajo F.J., Alcázar P., De Nuntiis P., Galán C. & Mandrioli P. 2006. May the definition of pollen season influence aerobiological results? *Aerobiologia* 22: 13–25.

Jones A.M. & Harrison R.M. 2004. The effects of meteorological factors on atmospheric bioaerosol concentrations — a review. *Sci. Total Environ.* 326: 151–180.

Kulmala M., Hämeri K., Aalto P.P., Mäkelä J.M., Pirjola L., Nilsson E.D., Buzorius G., Rannik U., Dal Maso M., Seidl W., Hoffmann T., Janson R., Hansson H.-C., Viisanen Y., Laaksonen A. & O'Dowd C.D. 2001. Overview of the international project on biogenic aerosol formation in the boreal forest (BIOFOR). *Tellus* 53B: 324–343.

Kulmala M., Suni T., Lehtinen K.E.J., Dal Maso M., Boy M., Reissell A., Rannik Ü., Aalto P., Keronen P., Hakola H., Bäck J., Hoffmann T., Vesala T. & Hari P. 2004. A new feedback mechanism linking forests, aerosols, and climate. *Atmos. Chem. Phys.* 4: 557–562.

Käpylä M. 1981. Diurnal variation of non-arboreal pollen in the air in Finland. *Grana* 20: 55–59.

Käärik A., Keller J., Kiffer E., Perreau J. & Reisinger O. Nilsson S.T. (eds.) 1983. *Atlas of airborne fungal spores in Europe.* Springer-Verlag, Berlin, Heidelberg, New York.

Laakso L., Hussein T., Aarnio P., Komppula M., Hiltunen V., Viisanen Y. & Kulmala M. 2003. Diurnal and annual characteristics of particle mass and number concentrations in urban, rural and Arctic environments in Finland. *Atmos. Environ.* 37: 2629–2641.

Lacey, M.E. & West J.S. 2006. *The air spora. A manual for catching and identifying airborne biological particles.* Springer, Dordrecht.

Lahti K. & Rönkä A. 2006. *Biologia: ympäristöekologia.* WSOY, Helsinki.

Lappalainen H.K., Sevanto S., Bäck J., Ruuskanen T.M., Kolari P., Taipale R., Rinne J., Kulmala M. & Hari P. 2009. Day-time concentrations of biogenic volatile organic compounds in a boreal forest canopy and their relation to environmental and biological factors. *Atmos. Chem. Phys.* 9: 5447–5459.

Lim D.V., Simpson J.M., Kearns E.A. & Kramer M.F. 2005. Current and developing technologies for monitoring agents of bioterrorism and biowarfare. *Clin. Microbiol. Rev.* 18: 583–607.

Lo E. & Levetin E. 2007. Influence of meteorological conditions on early spring pollen in the Tulsa atmosphere from 1987–2006. *J. Allergy Clin. Immunol.* 119, doi:10.1016/j.jaci.2006.11.612.

Madelin T.M. 1994. Fungal aerosols — a review. *J. Aerosol. Sci.* 25: 1405–1412.

Miguel A.G., Taylor P.E., House J., Glovsky M.M. & Flagan R.C. 2006. Meteorological influences on respirable fragment release from Chinese elm pollen. *Aerosol Sci. Technol.* 40: 690–696.

Morris C.E., Georgakopoulos D.G. & Sands D.C. 2004. Ice nucleation active bacteria and their potential role in precipitation. *J. Phys. IV France* 121: 87–103.

Morris C.E., Conen F., Huffman J.A., Phillips V. & Pöschl U. 2014. Bioprecipitation: a feedback cycle linking Earth history, ecosystem dynamics and land use through biological ice nucleators in the atmosphere. *Global. Change Biol.* 20: 341–351.

Mäkinen Y. 1981. Random sampling in the study of microscopic slides. *Rep. Aerobiol. Lab. Univ. Turku* 5: 27–43.

Möhler O., DeMott P.J., Vali G. & Levin Z. 2007. Microbiology and atmospheric processes: the role of biological particles in cloud physics. *Biogeosciences* 4: 1059–1071.

Nilsson S., Joseph Praglowski J. & Nilsson L. 1977. *Atlas of airborne pollen grains and spores in northern Europe*. Natur och kultur, Stockholm.

Norros V., Rannik Ü., Hussein T., Petäjä T., Vesala T. & Ovaskainen O. 2014. Do small spores disperse further than large spores? *Ecology* 95: 1612–1621.

Ogden E.C., Hayes J.V. & Raynor G.S. 1969. Diurnal patterns of pollen emission in *Ambrosia*, *Phleum*, *Zea*, and *Ricinus*. *Am. J. Bot.* 56: 16–21.

Phillips R. 2006. *Mushrooms. A comprehensive guide to mushroom identification*. MacMillan Ltd., London.

Prenni A.J., Petters M.D., Kreidenweis S.M., Heald C.L., Martin S.T., Artaxo P., Garland R.M., Wollny A.G. & Pöschl U. 2009. Relative roles of biogenic emissions and Saharan dust as ice nuclei in the Amazon basin. *Nature Geosci.* 2: 402–405.

Prenni A.J., Tobo Y., Garcia E., DeMott P.J., Huffman J.A., McCluskey C.S., Kreidenweis S.M., Prenni J.E., Pöhlker C. & Pöschl U. 2013. The impact of rain on ice nuclei populations at a forested site in Colorado. *Geophys. Res. Lett.* 40: 227–231.

Prospero J.P., Blades E., Mathison G. & Naidu R. 2005. Interhemispheric transport of viable fungi and bacteria from Africa to the Caribbean with soil dust. *Aerobiologia* 21: 1–19.

Pummer B.G., Bauer H., Bernardi J., Bleicher S. & Grothe H. 2012. Suspendable macromolecules are responsible for ice nucleation activity of birch and conifer pollen. *Atmos. Chem. Phys.* 12: 2541–2550.

Pöhlker C., Huffman J.A. & Pöschl U. 2012. Autofluorescence of atmospheric bioaerosols — fluorescent biomolecules and potential interferences. *Atmos. Meas. Tech.* 5: 37–71.

Pöhlker C., Huffman J. A., Förster J.-D. & Pöschl U. 2013. Autofluorescence of atmospheric bioaerosols: spectral fingerprints and taxonomic trends of pollen. *Atmos. Meas. Tech.* 6: 3369–3392.

Pöschl U., Martin S.T., Sinha B., Chen Q., Gunthe S.S., Huffman J.A., Borrmann S., Farmer D.K., Garland R. M., Helas G., Jimeney J.L., King S.M., Manzi A., Mikhailov E., Pauliquevis T., Petters M.D., Prenni A.J., Roldin P., Rose D., Schneider J., Su H., Zorn S.R., Artaxo P. & Andreae M.O. 2010. Rainforest aerosols as biogenic nuclei of clouds and precipitation in the Amazon. *Science* 329: 1513–1516.

Ranta H. & Satri P. 2007. Synchronized inter-annual fluctuation of flowering intensity affects the exposure to allergenic tree pollen in North Europe. *Grana* 46: 274–284.

Rantio-Lehtimäki A., Viander M. & Koivikko A. 1994. Airborne birch pollen antigens in different particle sizes.

Clin. Exp. Allergy 24: 23–28.

Reponen T. 1995. Aerodynamic diameters and respiratory deposition estimates of viable fungal particles in mold problem dwellings. *Aerosol Sci. Technol.* 22: 11–23.

Reponen T., Grinshpun S.A., Conwell K.L., Wiest J. & Anderson M. 2001. Aerodynamic versus physical size of spores: measurement and implication for respiratory deposition. *Grana* 40: 119–125.

Samson R.A., Houbraken J., Thrane U., Frisvad J.C. & Andersen B. (eds.) 2010. *Food and indoor fungi*. CBS Laboratory Manual Series 2, Utrecht, The Netherlands.

Schappi G.F., Taylor P.E., Pain M.C.F., Cameron P.A., Dent A.W., Staff I.A. & Suphioglu C. 1999. Concentrations of major grass group 5 allergens in pollen grains and atmospheric particles: implications for hay fever and allergic asthma sufferers sensitized to grass pollen allergens. *Clin. Exp. Allergy* 29: 633–641.

Schumacher C.J., Pöhlker C., Aalto P., Hiltunen V., Petäjä T., Kulmala M., Pöschl U. & Huffman J.A. 2013. Seasonal cycles of fluorescent biological aerosol particles in boreal and semi-arid forests of Finland and Colorado. *Atmos. Chem. Phys.* 13: 11987–12001.

Sesartic A., Lohmann U. & Storelvmo T. 2013. Modelling the impact of fungal spore ice nuclei on clouds and precipitation. *Environ. Res. Lett.* 8: 014029, doi:10.1088/1748-9326/8/1/014029.

Siljamo P., Sofiev M., Severova E., Ranta H., Kukkonen J., Polevova S., Kubin E. & Minin A. 2008. Sources, impact and exchange of early-spring birch pollen in the Moscow region and Finland. *Aerobiologia* 24: 211–230.

Simon-Nobbe B., Denk U., Poll V., Rid R. & Breitenbach M. 2008: The spectrum of fungal allergy. *Int. Arch. Allergy Immunol.* 145: 58–86.

Smith M., Emberlin J., Stach A., Rantio-Lehtimäki A., Caulton E., Thibaudon M., Sindt C., Jäger S., Gehrig R., Frenguelli G., Jato V., Rodriguez Rajo F.J., Alcázar P., Gálan C. 2009. Influence of the North Atlantic oscillation on grass pollen counts in Europe. *Aerobiologia* 25: 321–332.

Sofiev M., Siljamo P., Ranta H. & Rantio-Lehtimäki A. 2006. Towards numerical forecasting of long-range air transport of birch pollen: theoretical considerations and a feasibility study. *Int. J. Biometeorol.* 50: 392–402.

Spracklen D.V., Carslaw K.S., Merikanto J., Mann G.W., Reddington C.L., Pickering S., Ogren J.A., Andrews E., Baltensperger U., Weingartner E., Boy M., Kulmala M., Laakso L., Lihavainen H., Kivekäs N., Komppula M., Mihalopoulos N., Kouvarakis G., Jennings S.G., O'Dowd C., Birmili W., Wiedensohler A., Weller R., Gras J., Laj P., Sellegri K., Bonn B., Krejci R., Laaksonen A., Hamed A., Minikin A., Harrison R.M., Talbot R. & Sun J. 2010. Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation. *Atmos. Chem. Phys.* 10: 4775–4793.

Tack A.J.M., Hakala J., Petäjä T., Kulmala M. & Laine A.-L. 2014. Genotype and spatial structure shape pathogen dispersal and disease dynamics at small spatial scales. *Ecology* 95: 703–714.

Taylor P.E., Flagan R.C., Miguel A.G., Valenta R. & Glovsky

M.M. 2004. Birch pollen rupture and the release of aerosols of respirable allergens. *Clin. Exp. Allergy* 34: 1591–1596.

Tilak S.T. 1989. *Air borne pollen and fungal spores*. Vaijanti Prakashan, Aurangabad.

Tobo Y., Prenni A.J., DeMott P.J., Huffman J.A., McCluskey C.S., Tian G., Pöhlker C., Pöschl U. & Kreidenweis S.M. 2013. Biological aerosol particles as a key determinant of ice nuclei populations in a forest ecosystem. *J. Geophys. Res. Atmos.* 118: 10100–10110.

Vaitilingom M., Deguillaume L., Vinatier V., Sancelme M., Amato P., Chaumerliac N. & Delort A.M. 2013. Potential impact of microbial activity on the oxidant capacity and organic carbon budget in clouds. *Proc. Natl. Acad. Sci. USA* 110: 559–564.

Waggoner P.E. 1983. The aerial dispersal of the pathogens of plant-disease. *Phil. Trans. Royal Soc. London B* 302: 451–462.

Weber R. 2012. Impact of climate change on aeroallergens. *Ann. Allergy Asthma Immunol.* 108: 294–299.

Williams J., Crowley J., Fischer H., Harder H., Martinez M., Petäjä T., Rinne J., Bäck J., Boy M., Dal Maso M., Hakala J., Kajos M., Keronen P., Rantala P., Aalto J., Aaltonen H., Paatero J., Vesala T., Hakola H., Levula J., Pohja T., Herrmann F., Auld J., Mesarchaki E., Song W., Yassaa N., Nölscher A., Johnson A.M., Custer T., Sinha V., Thieser J., Pouvesle N., Taraborrelli D., Tang M.J., Bozem H., Hosaynali-Beygi Z., Axinte R., Oswald R., Novelli A., Kubistin D., Hens K., Javed U., Trawny K., Breitenberger C., Hidalgo P.J., Ebben C.J., Geiger F.M., Corrigan A.L., Russell L.M., Ouwersloot H.G., Vilà-Guerau de Arellano J., Ganzeveld L., Vogel A., Beck M., Bayerle A., Kampf C.J., Bertelmann M., Köllner F., Hoffmann T., Valverde J., González D., Riekkola M.-L., Kulmala M. & Lelieveld J. 2011. The summertime boreal forest field measurement intensive (HUMPPA-COPEC-2010): an overview of meteorological and chemical influences. *Atmos. Chem. Phys.* 11: 10599–10618.

Womack A.M., Bohannan B.J.M. & Green J.L. 2010. Biodiversity and biogeography of the atmosphere. *Phil. Trans. Royal Soc. B* 365: 3645–3653.

Yli-Panula E., Fekedulegn D.B., Green B.G. & Ranta H. 2009. Analysis of airborne *Betula* pollen in Finland, a 31-year perspective. *Int. J. Environ. Res. Public Health* 6: 1706–1723.

Appendix 1. Size ranges (μm) from literature and monthly mean concentrations (m^{-3}) of pollen grains, measured with the Burkard spore trap in Hyytiälä between 7 March and 29 August 2003.

	Mean size (μm)[a]	Mar	Apr	May	Jun	Jul	Aug
Alnus (alder), Betulaceae	18–21 × 23–30[b]	3	19	0	0	0	0
Artemisia (mugwort), Asteraceae	18–20 × 20–23[b]	0	0	0	0	1	0
Betula (birch), Betulaceae	19–26 × 21–30[b]	0	1	943	14	2	0
Chenopodium (goosefoot), Amaranthaceae	25–30[b]	0	0	0	0	0	0
Corylus (hazel), Betulaceae	20–25 × 25–28[b]	0	0	0	0	0	0
Poaceae (grasses)	24–43[b,c]	0	0	0	0	8	1
Cupressaceae; mainly *Juniperus* (juniper)	24–30[b]	0	0	4	15	1	0
Picea (spruce), Pinaceae	90–110[b]	0	0	4	1	0	0
Pinus (pine), Pinaceae	65–85[b]	0	1	8	250	6	1
Populus (aspen, poplar), Salicaceae	28–34[b]	0	1	2	0	0	0
Quercus (oak), Fagaceae	24–30 × 23–30[b]	0	0	0	0	0	0
Rumex (sorrel), Polygonaceae	17–22 × 20–23[b]	0	0	0	0	1	0
Salix (willow), Salicaceae	19–21 × 20–27[b]	0	0	2	0	0	0
Ulmus (elm), Ulmaceae	27–33 × 29–36[b]	0	0	0	0	0	0
Urtica (nettles), Urticaceae	13–17 × 15–20[b]	0	0	0	0	23	13
Total		3	22	962	282	45	17

[a] For elliptic or obovate grains two dimensions are given, while for spherical grains only one dimension is given. For Pinaceae grains with two laterally-placed bladders, the longest axis, sacci included, was measured.
[b] from Nilsson *et al.* (1977).
[c] No cereal pollen with larger grain size was observed.

Appendix 2. Pollen grain monthly mean concentrations (m^{-3}) measured with the Burkard spore trap in Hyytiälä between 3 March and 30 September 2004.

|	Mar	Apr	May	Jun	Jul	Aug	Sep
Alnus (alder), Betulaceae	0	66	1	0	0	0	0
Artemisia (mugwort), Asteraceae	0	0	0	0	1	4	0
Betula (birch), Betulaceae	0	14	189	3	0	0	0
Chenopodium (goosefoot), Amaranthaceae	0	0	0	0	0	0	0
Corylus (hazel), Betulaceae	0	0	0	0	0	0	0
Poaceae (grasses)	0	0	0	1	33	3	0
Cupressaceae; mainly *Juniperus* (juniper)	0	1	1	12	0	0	0
Picea (spruce), Pinaceae	0	0	2	0	0	0	0
Pinus (pine), Pinaceae	0	0	1	163	0	0	0
Populus (aspen, poplar), Salicaceae	0	11	2	0	0	0	0
Quercus (oak), Fagaceae	0	0	0	0	0	0	0
Rumex (sorrel), Polygonaceae	0	0	0	1	2	1	0
Salix (willow), Salicaceae	0	7	15	8	0	0	0
Ulmus (elm), Ulmaceae	0	0	0	0	0	0	0
Urtica (nettles), Urticaceae	0	0	0	1	26	21	0
Total	1	100	214	189	66	29	1

Appendix 3. Sizes (µm) from literature and monthly mean concentrations (m⁻³) of spores and other particles of biological origin measured with the Burkard spore trap in Hyytiälä between 6 March and 31 December 2003. n.a. = not available

	Size (µm)[a]	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Fungal spores: mitosporic [Deuteromycota (mould, mildew)]											
Alternaria	7–80 × 3–15[b]	2	2	5	4	25	14	13	0	0	0
Botrytis (grey mould)	8–14 × 6–9[b]	0	1	0	2	7	21	5	1	0	0
Cladosporium	3–25 × 3–9[b]	51	113	1134	587	5106	3578	2042	179	73	33
Epicoccum	15–25[b]	1	2	3	4	5	8	10	1	1	1
Erysiphaceae (powdery mildew)	19–25 × 10–14[c]	0	0	1	0	4	4	0	0	0	0
Helminthosporium sensu stricto	35–95 × 11–18[c]	0	0	1	1	3	4	3	0	0	0
Uredinales (stem rust); aeciospores and uredospores	16–40 × 13–24[d]	0	2	128	21	98	223	83	4	0	3
Ustilaginaceae (smuts); chlamydospores	4–16[d] (18–24[d])	0	2	9	36	144	98	9	4	5	6
Unidentified	n.a.; mainly < 10	52	76	1159	3173	4581	5421	4303	1466	50	55
Fungal spores: meiosporic [Basidiomycota (mushrooms); basidiospores]											
Suillus	2.5–4 × 7–10[d]	0	0	2	9	90	2825	7530	7	0	0
Boletus	5–6 × 15–19[d]	0	0	0	19	110	2020	1010	8	1	1
Russulaceae	5.5–8.8 × 6–10.5[d]	0	0	0	1	7	3604	2905	454	1	0
Unidentified	n.a.; mainly < 10	5	11	2087	4595	2976	7131	14508	9657	1046	138
Ascomycota (sac fungi); ascospore		48	137	3283	8662	7310	3736	1717	1136	411	124
Non-fungal spores and particles											
Myxomycota (slime molds)	6–12[d]	1	1	5	28	75	1347	517	0	3	0
Bryopsida (mosses) and Pteridophyta (ferns)	ca. 9–58[e,f]	1	1	6	7	11	13	7	0	0	0
Equisetum (horsetail)	35–40[f]	0	0	1	0	0	0	0	0	0	0
Part of mycelia	n.a.	0	0	11	31	38	39	60	4	5	13
Algae	ca. 10[f]	0	0	0	1	1	2	8	0	0	0
Nematoda (round worms)	n.a.	1	0	25	13	15	15	3	13	1	1
Unidentified	n.a.	0	1	0	1	0	0	2	0	0	0
Total		169	347	7863	17195	20604	30102	34538	12935	1595	375

[a] For elliptic or elongated spores in two dimensions are given, while for spherical spores only one dimension is given.
[b] from Samson *et al.* (2010).
[c] from Tilak (1989).
[d] from Käärik *et al.* (1983).
[e] from Lacey & West (2006).
[f] from Nilsson *et al.* (1977).

Appendix 4. Monthly mean concentrations (m^{-3}) of spores and other particles of biological origin, measured with the Burkard spore trap in Hyytiälä between 3 March and 4 August 2004. Note that values for August are not representative as the measurements ended on 4 August 2004.

	Mar	Apr	May	Jun	Jul	Aug
Fungal spores: mitosporic						
[Deuteromycota (mould, mildew)]						
Alternaria	0	1	6	11	17	52
*Botrytis (*grey mould)	0	0	3	10	29	57
Cladosporium	17	47	360	1097	2140	4885
Epicoccum	1	1	1	1	2	2
Erysiphaceae (powdery mildew)	0	0	2	0	0	0
Helminthosporium sensu stricto	0	0	0	0	0	2
Uredinales (stem rust); aeciospores and uredospores	0	1	45	17	46	107
Ustilaginaceae (smuts); chlamydospores	0	3	38	9	13	46
Unidentified	32	114	1270	1514	2857	3615
Fungal spores: meiosporic						
[Basidiomycota (mushrooms); basidiospores]						
Suillus	0	0	0	0	97	138
Boletus	0	0	0	12	483	881
Russulaceae	0	0	1	2	37	269
Unidentified	5	69	1890	10005	22326	35538
Ascomycota (sac fungi); ascospore[h]	36	359	3834	13151	13686	19657
Non-fungi spores and particles						
Myxomycota (slime molds)	0	13	16	13	102	52
Bryopsida (mosses) and Pteridophyta (ferns)	1	12	14	11	7	31
Equisetum (horsetail)	0	0	0	0	0	0
Part of mycelia	0	0	20	22	22	34
Algae	0	1	1	1	3	2
Nematoda (round worms)	0	3	64	151	85	67
Unidentified	1	0	0	0	2	0
Total	93	625	7563	26026	41958	65431
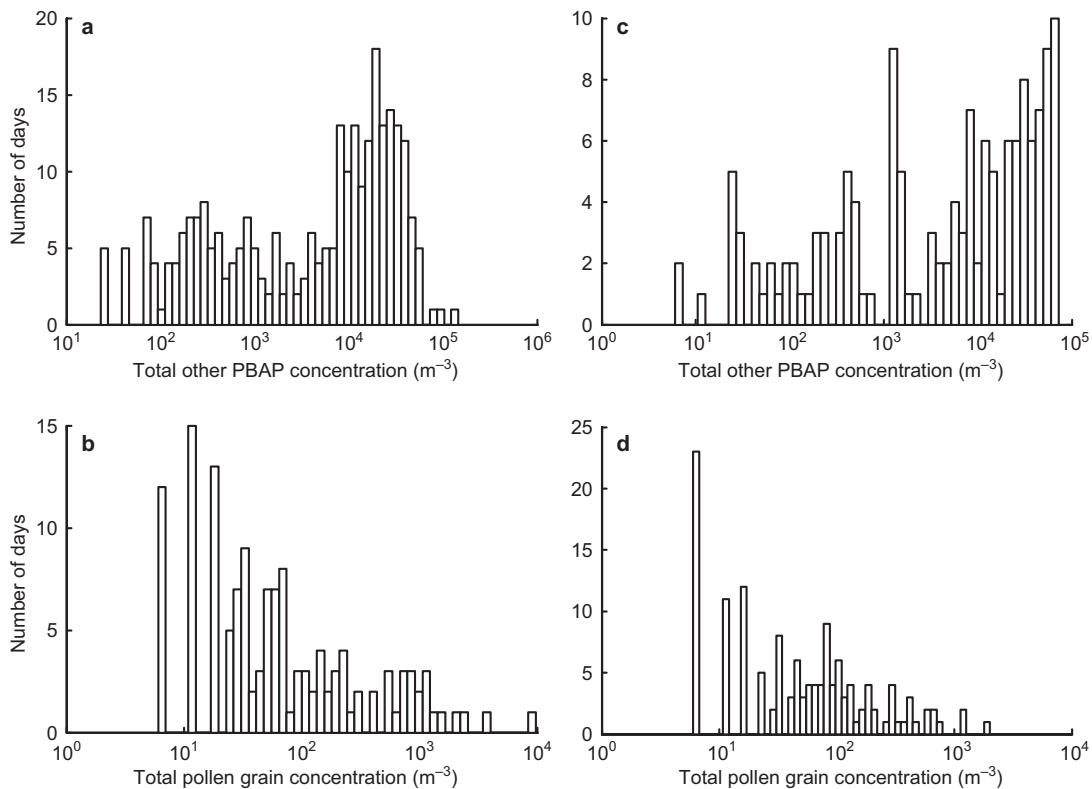
Appendix 5. Numbers of days when particular concentrations of other PBAP (**a**, **c**) and pollen (**b**, **d**) were recorded in 2003 (**a**, **b**) and 2004 (**c**, **d**).