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Seasonal cycles of fluorescent biological aerosol particles in boreal and semi-arid forests of Finland and Colorado

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Abstract

Biological aerosol particles have become increasingly important for atmospheric study, but continuous measurements at high time and size resolution have not been available until recently. Here we report seasonal cycles of fluorescent biological aerosol particles (FBAP) from the boreal forest in Hyytiälä, Finland (18 months) and the semi-arid Manitou Experimental Forest, Colorado (10 months). FBAP at both locations were observed to be highest in summer and lowest in winter, increasing by factors of 12 and 5 between these seasons, respectively. In addition to the low temperatures and reduced sunlight during winter, we suggest that snow cover inhibited FBAP release from local terrestrial surfaces and that more extensive snow cover at the Finland site contributed to lower winter FBAP concentrations. Average size distributions at each site exhibited peaks between 1.5 and 6 µm in aerodynamic diameter. The Finland site consistently showed a dominant, narrow FBAP peak at ~ 3 µm in addition to discreet modes at ~ 1.5 and ~ 5 µm, whereas the Colorado site showed broader peaks at 1.5 and 5 µm, suggesting different modes of biological particles at the two sites. FBAP concentrations in both locations were shown to correlate with daily patterns of relative humidity (RH) during each season. Also during summer at each site, average FBAP concentration scaled with RH, but at the Finland site RH values above ~ 82 % led to a significant decrease in FBAP concentration. We hypothesize that this is due to dew formation that inhibits bioparticle release. Lastly we show that rain during summer at each location led to pronounced increases in both fluorescent and total particle concentrations with FBAP peak particle size at ~ 2 µm and concentration scaling with rain intensity. We suggest that these particles are primarily fungal spores and other bioparticles lofted from splashing of rain droplets hitting soil and leaf surfaces. During the summer at the Colorado site we consistently observed a mode of ~ 4 µm particles appearing several hours after rain events that we suggest are fungal spores actively emitted when ambient conditions are most advantageous for spread and germination. The pronounced patterns of fluorescent bioparticles observed here suggest that parameterizations of

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both daily and seasonal cycles will be important to accurately reflect bioparticle emissions in future studies of atmospheric bioaerosols and their potential effects on clouds and precipitation.

1 Introduction

Biogenic gases and particles are released into the atmosphere from every region and ecosystem of the planet and contribute significantly to many Earth, atmospheric, and human systems. Small particles of biological origin (e.g. pollen grains, spores of fungi and plants, bacteria, and cellular debris) emitted directly into the atmosphere, called primary biological aerosol particles (PBAP), can range in size from approximately 0.1 to 100 µm and may be suspended for minutes to days (Womack et al., 2010; Després et al., 2012). While global estimates are uncertain, reports suggest that classes of PBAP exist in typical ambient concentrations of $\sim 10^4\ m^{-3}$ and often represent tens of percent of coarse particle number (Després et al., 2012 and references therein). PBAP not only play important roles in local and regional environments, but have also been observed after atmospheric transport of thousands of kilometers over land and oceans (Griffin et al., 2007; Polymenakou et al., 2008; Burrows et al., 2009; Hallar et al., 2011), and can sometimes utilize the atmosphere as a suspended habitat for multiple generations (Womack et al., 2010). Bioparticles may also influence climate forcing as airborne ice nuclei (e.g. Morris et al., 2004; Möhler et al., 2007; DeMott et al., 2010), especially in pristine regions which lack significant anthropogenic influence (Prenni et al., 2009; Pöschl et al., 2010). Bacteria known to be high temperature ice nucleators are ubiquitously observed in snow, rain, and fog water (Christner et al., 2008; Morris et al., 2008), and have also be observed in-situ in clouds well a.g.l. (Pratt et al., 2009; DeLeon-Rodriguez et al., 2013). Thus, because the role of clouds in the global energy balance is so uncertain (IPCC, 2007), a better understanding of the concentrations and properties of PBAP may help constrain aspects of climate uncertainty (Hoose et al., 2010; Sesartic et al., 2011). Ice-active bioparticles have been observed to increase

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dramatically during rainfall (Huffman et al., 2013; Prenni et al., 2013; Tobo et al., 2013) and may influence precipitation down-wind of particle sources, such as plants that host ice-active bacteria or spores. PBAP could thus influence precipitation patterns through human- or nature-induced landscape changes (Morris et al., 2013) and can also be inhaled into the lungs or deposited in the nasal passages, causing respiratory disease, allergies, and other health effects (Franze et al., 2005).

Despite the many systems that airborne bioparticles influence, detailed information about ambient PBAP patterns are missing, due largely to the fact that methods to detect and characterize them have been time consuming, costly, and prone to experimental bias. In particular, available techniques have traditionally been unable to detect PBAP with high time resolution or in narrow size fractions, and so time-resolved details of PBAP size, concentration, and source are poorly understood. Recent improvements in real-time measurement techniques have provided opportunities to monitor airborne bioparticles continuously at considerably higher frequency and lower cost. Field-portable techniques involving laser/light-induced fluorescence (LIF) have been particularly effective at rapidly providing information about PBAP in real-time (e.g. Hairston et al., 1997; Kaye et al., 2005; Sivaprakasam et al., 2009; Pan et al., 2011). Two commercially available LIF biosensors have been applied to ambient bioparticle monitoring and have helped to reveal fine detail about atmospheric PBAP patterns not previously observed (Gabey et al., 2010; Huffman et al., 2010; Toprak and Schnaiter, 2013). The ultraviolet aerodynamic particle sizer (UV-APS; TSI, Shoreview, MN, USA) and the wideband integrated bioaerosol sensor (WIBS; Droplet Measurement Technologies, Boulder, CO, USA) both characterize biological particles in real-time based on the intensity of fluorescence emission from individual particles after pulsed excitation at wavelengths characteristic for biofluorophores (Hill et al., 2009; O'Connor et al., 2011; Pöhlker et al., 2012). The UV-APS utilizes a single wavelength of excitation at 355 nm to induce fluorescence from molecules in the interrogated particle, including products of cellular metabolism (e.g. coenzymes NAD(P)H and riboflavin) among other biofluorophores (Eng et al., 1989; Pöhlker et al., 2012). Like all bioparticle detection

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techniques, real-time LIF instruments are not selective for all types of PBAP and thus have certain instrument-specific biases in their ability to classify particles as biological (e.g. Bones et al., 2010; Huffman et al., 2010; Gabey et al., 2013). Despite the uncertainty, fluorescent biological particles (FBAP) detected by the UV-APS or WIBS have been considered to be a lower-limit proxy for PBAP, with the understanding that certain classes of particles may be detected with less sensitivity (Huffman et al., 2010). This assumption was shown to be appropriate for remote Amazonian air when comparing UV-APS results with those from optical and electron microscopy samples collected simultaneously (Pöschl et al., 2010; Huffman et al., 2012), though additional studies of this type will be necessary to test the assumption under a wider variety of conditions.

Studies of seasonal bioaerosol variability have been performed and reviewed previously (Jones and Harrison, 2004). Long-term studies involving real-time LIF techniques, and thus providing high time and size resolution to FBAP detection, have only recently been attempted, however (Toprak and Schnaiter, 2013). Seasonal PBAP studies are important, because environmental factors that determine PBAP emission follow clear seasonality specific to ecosystem, and thus it is expected that airborne bioparticle patterns also follow seasonal patterns related as a function of microorganism species. Without detailed, long-term measurements, the ability to correctly model atmospheric effects of PBAP is significantly diminished. Here we compare continuous UV-APS measurements from two rural, forested sites: 18 months (2009–2011) in Hyytiälä, Finland and 10 months (2011–2012) near Woodland Park, Colorado, USA. To our knowledge this study provides the first report of a full seasonal cycle of UV-APS measurement data in the scientific literature and is the first to compare annual cycles of fluorescence bioparticle measurements from two geographic locations.

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2 Methods

2.1 UV-APS operation

The UV-APS draws in ambient aerosol and utilizes the time of particle flight between two red lasers (633 nm) to measure particle aerodynamic diameter (D_a) in 52 channels between 0.5 and 20 µm. A third laser (355 nm) is pulsed at each individual particle, and the intensity of the resultant fluorescence in the wavelength range of 420–575 nm (single channel; not wavelength-dispersed) is recorded (Hairston et al., 1997; Brosseau et al., 2000). Though particles as small as ∼ 0.3 µm can be detected, the particle collection efficiency drops below unity at approximately 0.8 µm. Thus, particle distributions that appear to peak at ∼ 0.8 µm are likely to be the tail of a larger mode (i.e. accumulation mode) peaking at smaller sizes. In addition, the influence of fluorescence from non-biological particles has been suggested to occasionally increase at small particle sizes (Huffman et al., 2010). For these reasons we chose 1.0 µm as the lower limit for integrated coarse particle numbers (N_c). We also utilize subscripts T and F to refer to total and fluorescent particles in the super-micron size range, respectively (e.g. $N_{T,c}$ and $N_{F,c}$). The upper limit on detectable particle size is practically defined by inlet engineering, but is limited to ∼ 20 µm by the UV-APS. Samples of ambient air were drawn through a total suspended particle (TSP) inlet head in each location and into the instrument at 5 L min^{-1}, where flow was split into 1 L min^{-1} of sample flow and 4 L min^{-1} of filtered sheath flow. The sampling frequency was every 5 min, with 285 s used for sample integration and ∼ 15 s used for data transfer from the UV-APS board to the dedicated computer (Huffman et al., 2010).

2.2 Finland sampling

The UV-APS was operated from inside the aerosol cottage at the SMEAR-II (Station for Measuring Ecosystem-Atmosphere Relations-II, 181 m elevation, lat. 61.85° N, long. 24.17° E) (Hari and Kulmala, 2005) in Hyytiälä, Finland from 27 August 2009 to

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17 April 2011 (598 days, 164 989 total 5 min samples). The sampling site is in a 51 yr old Scots pine forest and has been used extensively for atmospheric monitoring and has been described in detail elsewhere[1]. The TSP inlet utilized for this study was placed approximately 4 m above ground-level and 1 m above the cottage roof. The total flow through this inlet (10 L min^{-1}) was split between the UV-APS and a standard APS after being drawn vertically down through stainless steel tubing (OD ~ 0.75 in). The APS flow passed directly, and the UV-APS flow split at a ~ 45° angle via *y*-splitter and was brought to the instrument through ~ 0.7 m conductive rubber tubing (Simolex Rubber Corp., Plymouth, MI). New data files were initiated approximately every 7 days to keep the number of recorded 5 min samples per file manageable. The cottage was temperature-regulated during winter, and the inlet tube was mildly heated during warm months with a wrapped heating tape to prevent water vapor condensation upon entering the cooler lab.

2.3 Colorado sampling

The Colorado experiment took place within the Manitou Experimental Forest (2370 m elevation, lat. 39.10° N, long. 105.09° W), located approximately 48 km northwest of Colorado Spring, Colorado in a rural, semi-arid region of the central Rocky Mountains (2290 m a.s.l.). The closest town (Woodland Park) is located 15 km to the south. The vegetation surrounding the site is mixed ponderosa pine forest representative of large areas of the North American West (Ortega et al., 2013)[2]. Sampling was initiated as a part of the BEACHON-RoMBAS (Bio-hydro-atmosphere interactions of Energy, Aerosols, Carbon, H_2O, Organics and Nitrogen – Rocky Mountain Biogenic Aerosol Study) intensive field campaign July–August 2011 (Ortega et al., 2013)[3], and continued for almost a calendar year (20 July 2011–31 May 2012; 86 102 5 min sample points).

[1] http://www.atm.helsinki.fi/SMEAR

[2] http://web3.acd.ucar.edu/beachon/

[3] http://cires.colorado.edu/jimenez-group/wiki/index.php/BEACHON-RoMBAS

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The UV-APS was located in a climate-controlled (heating and cooling) trailer approximately 300 m from a local highway. A TSP inlet head was mounted approximately 2 m above the trailer roof, which was approximately 4 m above ground level. Stainless steel tubing (OD 0.75 in) ran vertically to an opening at the top of the trailer side wall. Approximately 1.5 m of electrically conductive rubber tubing (OD 0.75 in; Simolex) was connected to the vertical inlet tube and bent at a $\sim 30^{\circ}$ angle to pass into the trailer, then connecting to the vertical UV-APS instrument inlet piece. Data files were typically initiated every 1–3 days during the intensive BEACHON-RoMBAS campaign (20 July – 23 August 2011) and every 7–40 days for the rest of the sampling period.

2.4 Season definitions and ancillary instrumentation

For the long term comparisons between the two sites, we averaged time periods into seasons, as defined meteorologically: Spring (1 March–31 May), Summer (1 June–31 August), Fall (1 September–30 November), and Winter (1 December–29 February) (Trenberth, 1983).

Meteorological instruments measuring air temperature, relative humidity, barometric pressure, rain rate, wind speed, and wind direction are operated continuously at both sites and results were used for comparison. These instruments were located at 16.8 m above ground at the Finland site and 8 m above ground at the Colorado site.

3 Results and discussion

3.1 General trends

3.1.1 Particle concentration

The UV-APS was operated at each sampling location for a minimum of 10 months to evaluate the respective seasonal cycles of total and fluorescent biological particles. Though in very different sampling climates and environments, the seasonal trends at

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each location exhibit broadly similar patterns of higher FBAP concentrations in summer than in winter (Figs. 1–2, Table 1). FBAP at each site was highest May to October, and lowest December to February, however the relative increase between winter and summer was greater at the Finland site (factor of 12) than at the Colorado site (factor of 5). The Colorado measurements were performed for just less than one full seasonal cycle, whereas measurements were performed in Finland continuously for 18 months, allowing for comparison of late summer, fall, and winter seasons for two consecutive years. From this comparison (Fig. 1) the FBAP concentration data show fluctuations at weekly timescales or less, but are consistent between years at the seasonal level.

For example the magnitude of winter concentrations and the time period of decrease from elevated summer values (approx. mid-September) are each similar during the years of the Finland study. The low concentration of FBAP in winter is expected, due to the cold temperatures and snow cover in both locations, and is also similar to the general trend observed over a year-long study in SW Germany by Toprak and Schnaiter (2013). Environmental factors were expected to lower the concentration of airborne biological particles due to the reduced biological activity during winter and the reduced ability of microorganisms to become lofted to the air because of the snow coverage barrier. During the winter of 2011–2012, the Colorado site received ~ 120 cm of snow (D. Gochis, personal communication, 2013). However, during this period there were often long periods of dry, exposed ground, whereas the ground at the Finland site was continuously snow-covered from early November to mid-April (Fig. 1). This may partially explain the lower absolute concentrations in winter at the Finland site compared to the Colorado site, although the mean temperature is also lower and the latitude is higher at the Finland site. The FBAP dampening effect of low temperatures and snow cover in Finland is further suggested by a short period of relative increase in FBAP concentration during the late fall season 2009 (Fig. 1, green trace). Well after the FBAP concentration began to drop for the season in mid-September, and after the first few days of snow cover (9–16 November), approximately two weeks of warmer temperatures melted the snow and resulted in a marked increase in FBAP concentration. This

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short trend continued until a sharp decrease in temperature (1 December) caused a drop in FBAP by approximately 2 orders of magnitude. The total coarse aerosol concentration (Fig. S1) was unaffected by the steep temperature drop, however, and so the FBAP number fraction (Fig. S2) dropped precipitously and remained low until the beginning of spring 2010.

Seasonal bioaerosol measurements were previously performed at the Hyytiälä, Finland site, using a Burkard spore trap for sampling and optical microscopy for analysis, for part of two consecutive years (2003–2004) and show airborne fungal spore concentrations highest in summer and at lowest in winter (Manninen et al., 2008), consistent with observations reported here. Several other studies reporting measurements from rural Scandinavia and northern Asia have also reported similar seasonal trends, such as Kaarakainen et al. (2008) who showed fungi and bacteria concentrations from polymerase chain reaction (PCR) measurements were the highest in summer, also elevated in fall, and lowest in winter in central Finland. Makinen and Ollikainen (1973) discuss fungal spore concentrations peaking in summer and fall in an urban area of Finland, citing a number of previous studies showing various observations of seasonal trends for individual spore species. Matthias-Maser et al. (2000) show PBAP, using electron microscopy, peaking in summer at a remote Siberian sampling location. Thus, a number of studies have established the general trend of bioaerosol concentrations the highest during warm months at high latitudes. However, until recently the availability to investigate such trends at fine temporal resolution has not been available.

While FBAP concentrations show a seasonal trend peaking during warm months at both locations, trends for total coarse particles are distinctly different from those of FBAP. At the Finland site, no clear seasonal trend in total particle concentration was observable (Fig. S1a); mean concentration remained relatively constant across seasons. The total coarse particle number concentration in Colorado was the highest during spring and summer (Fig. S1b), in contrast to the FBAP trend, but the magnitude of increase from the lowest to the highest seasons was only a factor of ~ 2. Total particle number also exhibited significantly reduced daily and weekly variability as compared

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with FBAP (see vertical scatter in Figs. 1 and S1). The seasonal behavior of the ratio of FBAP to total coarse particles (Fig. S2) is similar to that of FBAP, showing that changes in the seasonal coarse aerosol composition are dominated by changes in FBAP and not by total particles. For example, it has been speculated that atmospheric dilution caused by the increasing boundary layer height might cause the often observed decrease in FBAP concentration during the day. Seasonal averages of particle concentration do not clearly support this suggestion, however, because concentrations of coarse particle in winter at both sites are similar to, or well below, summertime averages when the boundary layer height is expected to be much higher. Physical effects of atmospheric dilution would also be expected to treat biological and non-biological particles similarly, suggesting that the observed wintertime decreases in FBAP are indeed less related to atmospheric dilution than to changes in biological emission patterns. Further, the seasonally stable total coarse aerosol concentration at the Finland site, despite months of continuous snow coverage, suggests that the atmospheric source of total particles is not dominated by wind-blown re-suspension of soil or leaf litter and, by extension, that observed FBAP at this location is not primarily attached to lofted mineral dust particles. These observations thus suggest that boundary layer effects may contribute more weakly to FBAP trends than do biological factors, as also suggested by Huffman et al. (2012).

3.1.2 Size of biological particles

Size distributions of FBAP number concentrations reflect typical peaks between 1–7 µm in all cases reported here. However, while particle concentration trends were consistent between the two measurements sites, trends in particle size were less so, indicating differences in the bioaerosol sources between the sites. Season-average FBAP size distributions from the Finland site (Fig. 3) show relatively narrow peaks at approximately 2.5 µm and a shoulder at approximately 1.2 µm for each individual season averages of spring, summer, and fall. Though the peaks of the seasonally averaged size distributions are relatively narrow, they are comprised of and broadened

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by peaks of individual bioaerosol types at different sizes. This idea is highlighted by Fig. 4c that shows an example of size distributions average over a relatively short time period (~ 6 h) where discreet particle modes are resolved enough to see their influence to the average distribution.

Through the seasons sampled, particle modes were observed to vary significantly in particle size and concentration, but three modes were consistently observed, at: ~ 1.5, ~ 3, and ~ 5 µm, respectively, in addition to particles occasionally present < 1 µm. The mode at ~ 3 µm, however, was the most common individual mode, and was often observed to have extremely narrow distribution width. Further, the fraction of particles represented by each of these three individual modes observed during spring through fall was not obviously correlated with meteorological variables and is thought to have been influenced heavily by local biological activity. The FBAP modes observed here are consistent with results from a recent laboratory study revealing that the average aerodynamic spore size of 66 fungal species collected from around Hyytiälä to be between 1.5–5.1 µm (Hussein et al., 2013). Figure 5 shows an example of a period with the narrow FBAP mode that is consistent both in size and in concentration for several days at a time during the fall. The peak was commonly observed to be large enough to overwhelm the size distribution of total particles (Fig. 5a), a rare scenario among ambient UV-APS observations in general. The FBAP peak clearly influences the total particle distribution at that size, but $N_{F,c}$ only represents 51 % of $N_{T,c}$ at the peak of the distribution (Fig. 5b). Each particle mode has a distribution of particle size and fluorescence intensity, and the instrument detector sensitivity defines whether an interrogated particle will be counted as fluorescent. Huffman et al. (2012) showed that the UV-APS can sometimes undercount fluorescence from particles of a single mode so the technique will remain less affected by weakly fluorescent, non-biological particles. The evidence here supports the previous observation of a single mode containing a distribution of fluorescence intensity. Thus, the remaining 49 % of particles in the mode observed here (Fig. 5) exhibit fluorescent emission below the FBAP threshold, but are likely to be biological particles whose physiological or metabolic state has changed, leading

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to a change in fluorophore content (Wu and Warren, 1984a,b; Roshchina, 2003; Tack et al., 2013).

Despite the relative consistency during the other seasons, the winter average size distribution at the Finland site (Fig. 3d) is qualitatively different than the other three seasons, because the highest observed peak was at 1.2 µm, with the 2.5 µm peak as a large shoulder. Additionally, both the fall and winter averages show a small peak at 0.8 µm. This is a false "peak," however, caused by instrument collection efficiency that drops below approximately 0.8 µm. It is therefore impossible to know the shape of the distribution below this particle size from UV-APS data, but it is likely that these particles are the tail of a larger, accumulation mode peaking well below 1 µm (Dal Maso et al., 2005). With rare exception (e.g. Fig. 4c), these small particles were generally only observed in fall and winter. For example, when the temperatures are low, the atmospheric boundary layer is also usually lower, increasing the particle concentration within a smaller volume. Thus, these particles may be related to wood-burning emissions during cold days when the boundary layer is shallow and may be small pieces of ash, or PAH-containing particles that could fluoresce strongly (Aizawa and Kosaka, 2008).

In contrast to the Finland site, the trends in the average FBAP size distribution from the Colorado site show two clearly separated modes at approximately 1.5 and 5.0 µm, with the exception being summer when additional modes resulting from rain influence were most commonly observed (see Sect. 3.2.3), leading to a broad single peak. Also in contrast to the Finland site, the number of individual FBAP modes observed in Colorado at any given time was usually fewer, and season averages more closely approximate the qualitatively stable size distributions on any given day. The submicron FBAP mode was not observed at the Colorado site, possibly because it is more remote than the Hyytiälä station with respect to residential areas and thus has less influence from nearby wood burning.

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3.1.3 Diurnal patterns

One of the benefits of high-time resolution LIF bioaerosol techniques is the ability to analyze diurnal (daily or 24 h) averages of size distributions and total concentrations and monitor how these patterns change over time. Consistent to what has been reported previously (Huffman et al., 2010; Gabey et al., 2011, 2012; Toprak and Schnaiter, 2013) $N_{F,c}$ and $N_{F,c}/N_{T,c}$ at both the Finland and Colorado sites peak during every season in the evening or early morning hours when relative humidity (RH) is the highest and temperature is the lowest (see also Sect. 3.2). Seasonally averaged diurnal plots for the Finland site highlight the extremely narrow mode at ~ 3 µm (Fig. S3). Similar particle size distribution plots for the Colorado site (Fig. S4), however, show size distributions peaking at 2, 3, and 5 µm at different times of the night for different seasons. These patterns are discussed in more detail in the Supplement (SOM).

3.2 Meteorological effects on fluorescent bioparticles

3.2.1 Temperature effects

Over the course of the year and as a function of whole season averages, there is a positive correlation between temperature and $N_{F,c}$ (Fig. S5). Increasing the ambient temperature appears to increase the relative number of FBAP. These seasonal trends are misleading, however, because the trend as viewed on a daily basis (Figs. S3 and S4) shows that $N_{F,c}$ decreases with increasing temperature during the day. This suggests that biological processes that determine the release of bioaerosols are strongly a function of season and may require a certain minimum temperature to function. Spring recovery in connection with the increased photosynthetic activity of the biosphere has also been linked to nanoparticle formation (Dal Maso et al., 2009). Once the biological activity has been initiated, however, higher temperatures and lower RH during the day actually attenuate bioaerosol release. At the Finland site, $N_{F,c}$ appears relatively constant at ~ 0 cm^{-3} until the temperature in any season exceeds ~ 0 °C, likely due to snow

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melting, at which point $N_{F,c}$ increases with temperature (Fig S5a). At the Colorado site, the minimum temperature was not as consistent or definitive, but exceeding a threshold of approximately −5 °C appears necessary for $N_{F,c}$ to increase (Fig. S5b). Ambient temperature has been shown to influence fungal spore release, but this process is strongly a function of fungal species. Airborne fungi are highly diverse and some species (e.g. *Cladosporium*) favor release during warm, dry periods, whereas other spores (e.g. many basidiospores and ascospores) favor release during cooler, wet periods (De Groot, 1968; Gilbert, 2005; Elbert et al., 2007; Fröhlich-Nowoisky et al., 2009, 2012; Huffman et al., 2013).

3.2.2 Humidity effects

Air temperature correlates strongly, but inversely, with RH by affecting the saturation water vapor pressure in air. So it is no surprise that $N_{F,c}$ shows an opposite trend with RH as it does with temperature on a daily basis (e.g. Gregory and Hirst, 1957; Gottwald et al., 1997; Burch and Levetin, 2002; Elbert et al., 2007). However, in contrast to temperature, RH shows the same general correlation with $N_{F,c}$ on both daily and seasonal levels (Fig. 6, S3 and S4). At the Finland site (Fig. 6a) RH levels in winter are generally higher due to lower temperatures, but $N_{F,c}$ is almost continuously near zero, thus showing little correlation. During spring and fall the average $N_{F,c}$ concentration is higher, but the relationship between $N_{F,c}$ and RH is inconsistent. During summer there is a clearly positive correlation between $N_{F,c}$ and RH from ~ 30–82 % RH suggesting that many of the bioparticles detected could be ejected actively by RH-dependent mechanisms (Ingold, 1999; Pringle et al., 2005; Elbert et al., 2007). Above RH ~ 82 % during summer, however, $N_{F,c}$ decreases substantially. A similar trend was observed using UV-APS data from a remote site in Amazonia (Huffman et al., 2012), but was unexplained. Measurements of temperature and RH allow estimation of dew point for each sample, though surface temperature and RH immediately above vegetative surfaces may be somewhat different from measurements at 8 m above the forest floor. RH of ~ 82 % corresponds, on average, to a temperature ~ 2.7 °C above the dew point of water (Fig. S6). There-

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fore, we hypothesize that at this RH dew is beginning to form on plant and terrestrial surfaces, thus applying a layer of water through which spores and other bioparticles are unable to escape. This, in turn, lowers the FBAP concentration. It is also possible that this hypothesis could partially explain why RH lags behind $N_{F,c}$ in the diurnal cycle (Sect. S1.1, Figs. S3 and S4). Many species of fungi utilize increased RH overnight to promote active spore release, but the formation of dew when RH peaks may inhibit the ability of spores to overcome local the micro-scale boundary layer, and thus $N_{F,c}$ would begin to decrease in the morning as dew begins to form and before RH decreases.

The trends observed at the Finland site are broadly similar for the Colorado site: (1) $N_{F,c}$ is very low during winter, (2) during spring and fall $N_{F,c}$ is somewhat higher, but the relationship with RH is inconsistent, and (3) during summer the positive correlation between $N_{F,c}$ and RH is clear. The Colorado site shows a consistent, positive relationship between RH and FBAP concentration. The relationship of $N_{F,c}$ with RH could be important for modeling bioaerosol emission, and periods of strong correlation suggests that concentrations measured under these conditions were dominated by recent, local emission, rather than by long-range transport.

3.2.3 Precipitation effects

In addition to FBAP patterns associated with daily temperature and RH cycles, during some seasons we observed sudden, substantial increases in FBAP concentrations immediately upon arrival of rain. Such correlations scaled with rain intensity, with FBAP increases the strongest during heavy rain, but still observed during light drizzle. The relationship between bioparticles and rain has been reported previously (Faulwetter, 1917; Gregory and Hirst, 1957; Hirst and Stedman, 1963; Fitt et al., 1989; Constantinidou et al., 1990; Pinkerton et al., 1998; Allitt, 2000; Paul et al., 2004; Huffman et al., 2013; Prenni et al., 2013), and was especially apparent during the summer season at the Colorado site, when even weak or short rain events were observed to increase $N_{F,c}$ by a factor of 6–40 over the concentration present immediately before precipitation began. Rain also caused an increase in the total particle concentration, $N_{T,c}$, though by

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a smaller factor (about an average of 1.3×) than $N_{F,c}$. Huffman et al. (2013) discussed that increases in aerosol concentration did not correlate with changes in wind patterns before or during storm arrival, and submicron aerosol concentrations follow a different temporal pattern related to rain (Laakso et al., 2003). Microscopy images from samples collected during the Colorado measurement campaign suggest the majority of the increase in total particles was caused by bioparticles, despite UV-APS categorization as non-fluorescent (Huffman et al., 2013). Aerosol concentrations during and after rain events followed a relatively consistent and repeatable pattern throughout the summer and into the fall at the Colorado site (e.g. Fig. S7). Immediately upon arrival of rain, FBAP concentrations increased and corresponding size distributions shifted from peaking at ~ 3–4 µm to ~ 2.0 µm. Subsequent pulses of rain each resulted in $N_{F,c}$ increases. Several (~ 8) hours after the rain, however, a second mode peaking at 4.5 µm usually became apparent and stayed elevated for ~ 12 h in many cases (e.g. McCartney and Lacey, 1990). Figures 7 shows average FBAP size distributions for the Colorado site after separating into periods during rain, after rain, and without any rain influence. While this categorization is difficult due to the lack of discreet break-points between periods, and thus the averages are smoothed by cross-influence, the averaged periods highlight the differing mode size and concentration behaviors. Size distributions during rain-influenced periods show a narrow peak at 2.0 µm in size at higher FBAP concentration than the other two average periods shown. After-rain periods show influence from the ~ 2 µm particles that appear during rain, but also show a high concentration of a ~ 4 µm mode of particles with little tail. Periods without rain show a mode peaking at 4.1 µm, with broad width and featureless appearance suggesting a large mixture of particle types and sizes. Compared to periods without rain influence, the integrated $N_{F,c}$ increased on average by approximately factors of ~ 5 and ~ 2 during and after rain, respectively.

Rain events occurred commonly during the afternoon, thus strongly influence diurnal patterns (see Sect. S1.3, Fig. S8). We hypothesize that FBAP observed during rain is either being washed out of the column of air above the measurement site by falling

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rain drops and re-suspended when droplets hit the ground and shatter, or they are being released mechanically from plant and soil surfaces by the agitation of falling hydrometeors. We further hypothesize that the mode of particles commonly observed hours after the rain are spores being actively emitted by fungi taking advantage of moist, cool air for germination. In a related studies we reported that bioparticles during the BEACHON-RoMBAS study were also observed to be highly ice active (Huffman et al., 2013; Prenni et al., 2013; Tobo et al., 2013), suggesting that such bioparticles could influence ice cloud formation if lofted sufficiently.

At the Finland site, rain was also observed to cause a sudden increase in FBAP concentration. However, in contrast to observations at the Colorado site, no after-rain effect was observed in Finland and the magnitude of $N_{F,c}$ increase was considerably less than in Colorado. The reason behind this may be that actively ejected fungal species present in the relatively dry Colorado forests need to react quickly to elevated RH after rain for ecological advantage, whereas species in more humid environments, such as the Finnish forest, can be released more continuously without need for immediate fitness gain (e.g. Pinkerton et al., 1998). The observed pattern at the Finland site was that as rain began to fall, $N_{F,c}$ increased sharply, but quickly returned to approximate pre-rain levels (Fig. S9). The predominant particle mode during dry periods peaked at 2.9 µm, but during rain a 2.2 µm mode dominated. As with the observations in Colorado, the relationship between rain and FBAP increase was strongest in summer periods and was less consistent during other seasons.

4 Conclusions

Fluorescent biological aerosol particles were measured continuously in real-time at two forested sites using a UV-APS. The boreal site in Hyytiälä, Finland and the semi-arid site in the Manitou Experimental Forest, Colorado showed similar seasonal FBAP cycles, with concentrations highest during the summer (0.051 cm^{-3} and 0.030 cm^{-3}, respectively) and lowest during winter when temperatures were the coldest. Snow cover

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appeared to prevent local bioaerosol emission from vegetated surfaces, highlighted by correlation of snow cover measurements with winter FBAP concentrations lower in Finland than in Colorado, where dry ground was more often free of snow. Total particle number did not follow the same trend, remaining relatively constant throughout the year at each sampling location and showing minimal diurnal periodicity. These observations suggest that the source of fluorescent bioparticles was different from that of the non-fluorescent particles that dominated the total coarse particle number and that FBAP concentrations were influenced more heavily by biological emission than by boundary layer meteorology. Previous studies have suggested that certain bioparticles nucleate ice at temperatures sufficient to influence mixed-phase cloud formation and evolution (Möhler et al., 2007), and work has been done recently to globally model bioparticle effects on clouds (Hoose et al., 2010; Sesartic et al., 2011; Burrows et al., 2013). The strong cycles of FBAP observed here suggest that such parameterizations of bioparticle concentrations need both seasonal and daily temporal elements.

The FBAP at each site discussed here exhibited distinct trends, but was most concentrated between 1.5 and 6 µm in particle size, as has been reported previously. At the Finland site three FBAP modes were commonly observed, at: ∼ 1.5, ∼ 3, and ∼ 5 µm, with additional particles observed below the 1 µm considered here as the instrument cut-off. The 2.5–3.0 µm mode was nearly ubiquitous and often extremely narrow in width. This mode dominated the FBAP size distributions during spring, summer, and fall, but during the winter season the FBAP number distribution shifted to peak at ∼ 1.5 µm. The appearance and disappearance of most individual particle modes were not obviously explainable by changes in wind speed or wind direction, suggesting that particle sources were either not local in nature, or came from multiple nearby point sources. In contrast to the Finland site, FBAP number distributions at the Colorado site were dominated by modes at 1.5 µm and 5.0 µm throughout the observed seasons. Both sites also show the same daily trends as have been reported previously (Huffman et al., 2012; Toprak and Schnaiter, 2013), with FBAP concentrations generally following RH patterns of daily maxima during the late night and early morning and minima dur-

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ing the day. Hourly particle size distributions were considerably broader at the Colorado site than at the Finland site, however.

Air temperature, relative humidity and precipitation were observed to correlate strongly with FBAP concentrations at both sites. As viewed on a yearly or seasonal basis, higher temperatures were correlated with higher bioparticle concentrations, which is likely to reflect an increase of biological activity and bioparticle release with ambient temperature. On a daily basis, however, FBAP was generally lowest during the day when temperatures were highest and when relative humidities were lowest, as discussed below. We observed much lower FBAP concentrations during winter and when averaged seasonally, a minimum temperature of 0 or −5 °C was necessary at the Finland and Colorado sites, respectively, to initiate seasonal FBAP increase.

Relative humidity was also shown to correlate with FBAP concentration. During winter at both sites the increase of FBAP with RH was minimal due to the low temperatures, snow cover, and reduced biological activity. As with temperature, elucidating trends of the RH and FBAP relationship from seasonal averages is not always clear. During spring, summer, and fall at each site, FBAP increased with RH during the day, but did not necessarily correlate similarly as a season average. During summer at the Finland site, however, there was a clear, consistent FBAP increase with increasing RH, up to an RH value of 82 %, above which the FBAP concentration dropped steeply. We hypothesize that during the summer, increased RH at night triggers the active wet discharge of bioparticles such as fungal spores, but that this release is hindered by the formation of dew when the ambient temperature gets close to the dew point temperature. During the summer at the Colorado site, however, this effect was not observed, and FBAP increased steadily with RH.

Rain also played a significant role influencing FBAP concentrations at both sites. During summer and fall at the Colorado site, the rain caused a sharp spike in concentration of FBAP, peaking at 2.0 µm, that scaled with rain intensity and lasted minutes to hours. The relative increase of these particles overwhelmed even the total coarse particle concentration, which increased by a factor of 6–40 during rain. We suggest that

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rain causes an increase in FBAP by releasing bioparticles from the ground via some splash-based mechanism that ejects terrestrial spores and bioparticles (e.g. Faulwetter, 1917; Butterworth and McCartney, 1991; Madden, 1997; Huber et al., 1998; Morris et al., 2011). Several hours after the rain a particle mode peaking at 4.5 µm consistently became present and remained elevated for up to twelve hours while bioparticles of all other sizes dissipated. The pattern was repeatable during rain events through the summer and into the fall. At the Finland site we observed a similar increase of FBAP at 2 µm during rain. The relative increase during rain at the Finland site was weaker than at the Colorado site, however, with no after-rain period. Additionally the concentration of the dominant 3 µm mode usually present before rain was reduced.

Overall, the long-term measurement results reported in this study confirm that the emission and abundance of biological aerosol particles in forest air are closely linked to meteorological conditions. They are consistent with earlier studies suggesting a tight coupling between biological aerosols and the hydrological cycle, which may have an influence on the formation and properties of clouds and precipitation, and thus on regional and global climate.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/13/17123/2013/acpd-13-17123-2013-supplement.pdf.

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Table 1. Seasonal comparison of coarse-mode total aerosol particle ($N_{T,c}$), fluorescent biological aerosol particle ($N_{F,c}$) and bioparticle ratio ($N_{F,c}/N_{T,c}$) trends at Finland and Colorado sites, respectively. Particle concentrations shown as arithmetic mean values averaged over entire season. Variability in each case listed as ± standard deviation. Modes discussed represent most consistently observed peaks in particle size distribution, not necessarily corresponding to peaks in size distribution averages. Rain was not observed during winter season at either sampling location due to cold temperatures. Particle mode width listed as: b = broad, vb = very broad, n = narrow.

Hyytiälä, Finland					
		Spring	Summer	Fall	Winter
$N_{T,c}$ (cm^{-1})		0.43 ± 0.53	0.45 ± 0.39	0.41 ± 0.42	0.47 ± 0.34
$N_{F,c}$ (cm^{-1})		0.015 ± 0.024	0.046 ± 0.048	0.027 ± 0.032	0.004 ± 0.0046
$N_{F,c}/N_{T,c}$		0.044	0.13	0.098	0.011
Particle Modes (µm)	1			<1.0(n)	
	2	1.5(n)	1.5(n)	1.5(n)	1.5(b)
	3	3.0(b)	3.0(n)	3.0(n)	2.5(b)
Temperature (°C)		2.27 ± 7.44	16.26 ± 5.31	3.64 ± 6.66	−10.56 ± 6.41
Relative Humidity (%)		71.8 ± 20.0	60.92 ± 17.51	82.52 ± 12.17	97.05 ± 4.48
$N_{F,c}$ vs Rain (Correlation)		Moderate	Moderate	Minimal	None
Diurnal $N_{F,c}$ Peak (Hour of Day)		2:00	23:00	20:00	23:00
Manitou Experimental Forest, Colorado					
		Spring	Summer	Fall	Winter
$N_{T,c}$ (cm^{-1})		0.73 ± 0.72	0.44 ± 0.29	0.28 ± 0.23	0.20 ± 0.20
$N_{F,c}$ (cm^{-1})		0.015 ± 0.024	0.030 ± 0.030	0.017 ± 0.031	0.0053 ± 0.0063
$N_{F,c}/N_{T,c}$		0.025	0.088	0.057	0.030
Particle Modes (µm)	1	2.0(b)	2.0(vb)	1.5(b)	1.5(b)
	2	5.0(b)	4.5(vb)	5.0(b)	5.0(b)
Temperature (°C)		6.82 ± 8.46	17.68 ± 7.20	5.60 ± 8.57	−3.82 ± 7.52
Relative Humidity (%)		48.34 ± 30.03	51.89 ± 29.23	56.41 ± 27.57	56.92 ± 25.69
$N_{F,c}$ vs Rain (Correlation)		Moderate	Strong positive	Moderate	None
Diurnal $N_{F,c}$ Peak (Hour of Day)		23:00	22:00	3:00	22:00

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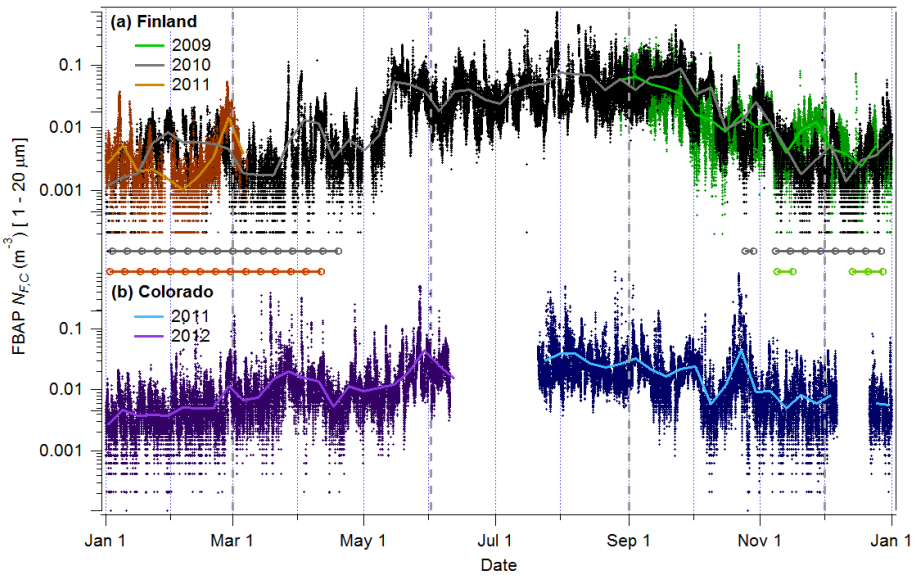

Fig. 1. Overview of FBAP concentration and key meteorological data at each site. Top traces represent 24 h mean values of ambient air temperature, plotted on right axes as shown. Small dots represent individual 5 min data points from UV-APS. Colored traces cutting through UV-APS data show 7 day mean values of FBAP concentration, plotted on left axes. Axis ranges matched in upper and bottom panels. Open circle markers represent periods of ground snow cover at Finland site (not available at Colorado site) and are shown for each recorded observations (approximately every week through winter season). Dashed vertical lines show seasonal boundaries used for averaging (as discussed in Sect. 2.4)

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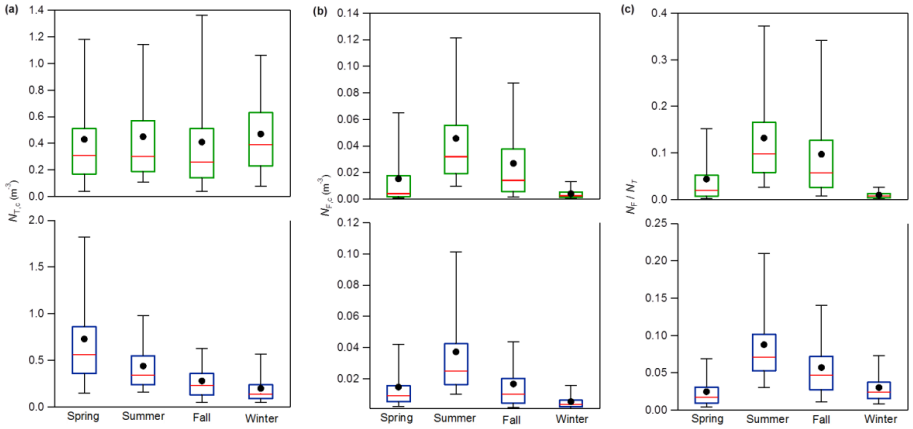

Fig. 2. Seasonal concentrations of coarse-mode total and fluorescent bioparticles. Finland site shown in upper panels as green boxes and for Colorado site in lower panels as blue boxes. Whisker plots represent mean and median values (black dots, red lines, respectively), 25th and 75th percentiles (boxes), 5th and 95th percentiles (vertical lines). **(a)** Concentrations of total particles, $N_{T,c}$. **(b)** Concentration of fluorescent bioparticles, $N_{F,c}$. **(c)** Ratio of fluorescent to total particles, $N_{F,c}/N_{T,c}$.

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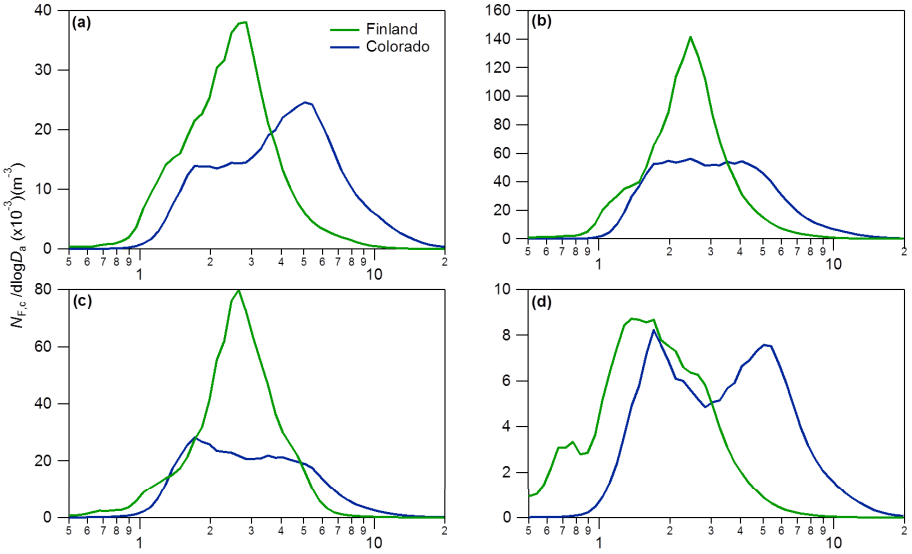

Fig. 3. Mean FBAP number distributions for: **(a)** Spring **(b)** Summer **(c)** Fall **(d)** Winter at Finland (green) and Colorado (blue) sites.

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Seasonal cycles of fluorescent biological aerosol particles

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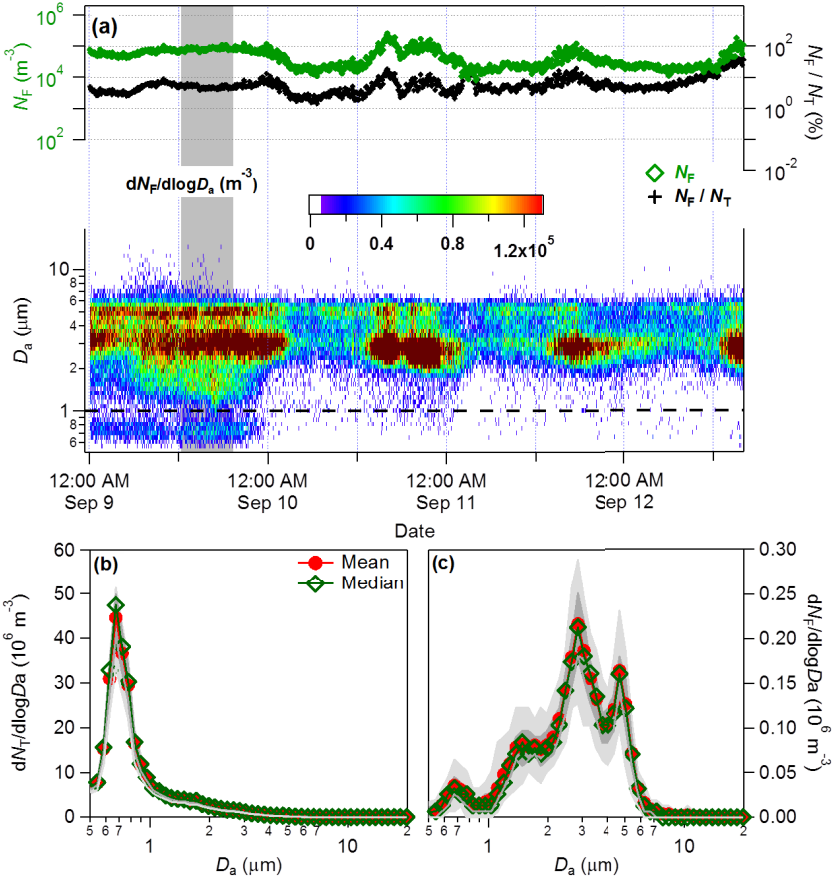

Fig. 4. Representative particle modes present at Finland site. **(a)** Image plot with gray bar highlighting period of average (14:56–21:06, 9 September 2009). Size distributions of: **(b)** N_T and **(c)** N_F. Mean and mean traces shown, with gray region showing inner quartile.

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Seasonal cycles of fluorescent biological aerosol particles

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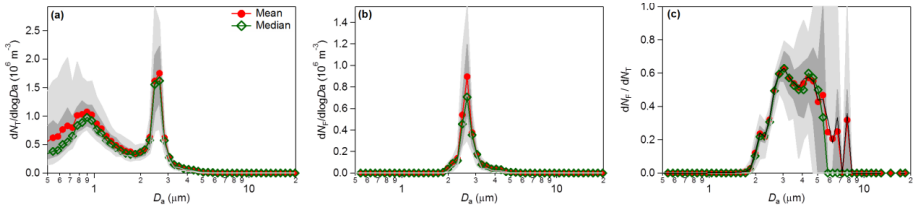

Fig. 5. Total particle and FBAP number size distributions from Finland site showing narrow FBAP peak at 2.64 μm. Average of samples from 8 October 2009 18:53–21:51, (35 total samples). Mean and median lines shown as red and green traces, respectively. Dark gray zone shows 25–75th percentiles, and light gray zone shows 5th–95th percentiles.

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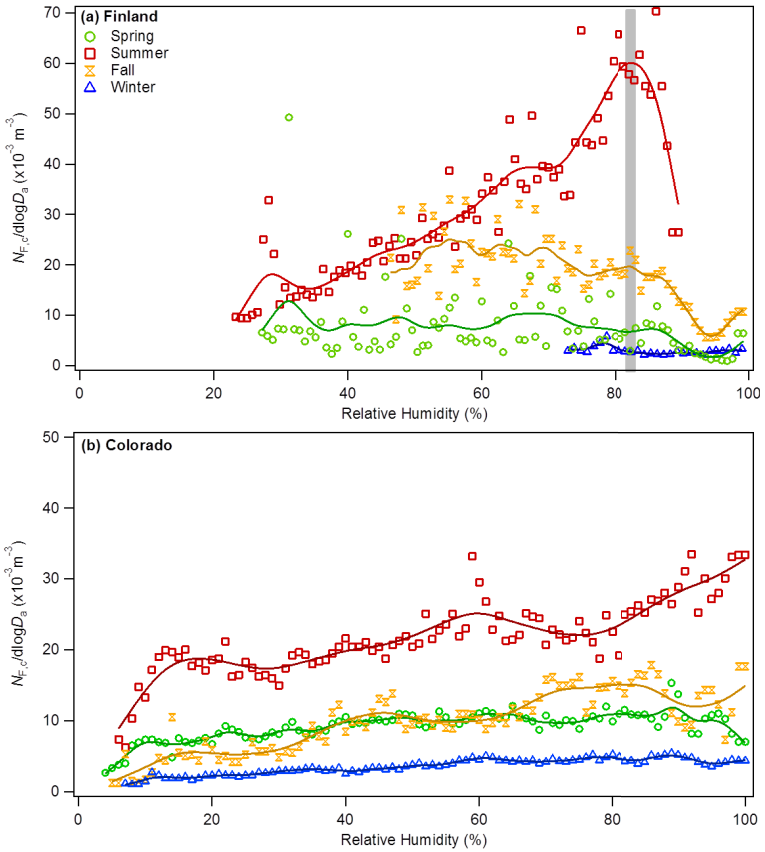

Fig. 6. Seasonal comparison of median bioparticle concentration to relative humidity at **(a)** Finland and **(b)** Colorado sites. Data averaged into 100 RH bins. Bins containing less than 0.02 % of the total points were removed. Fit lines are spline curves to guide the eye. Gray bar shows approximate RH value where FBAP concentration decreases.

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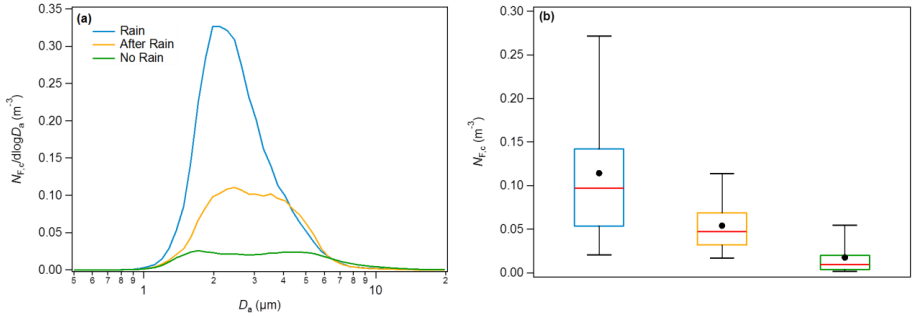

Fig. 7. Summer average rain influence in Colorado. **(a)** Mean size distributions of N_F concentration: during rain (blue), immediately after rain (yellow), and without rain (green). **(b)** Whisker plots showing $N_{F,c}$ concentration present: during rain (blue), immediately after rain (yellow), and without rain (green). Whisker plots represent mean and median values (black dots, red lines, respectively), 25th and 75th percentiles (boxes), 5th and 95th percentiles (vertical lines).

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