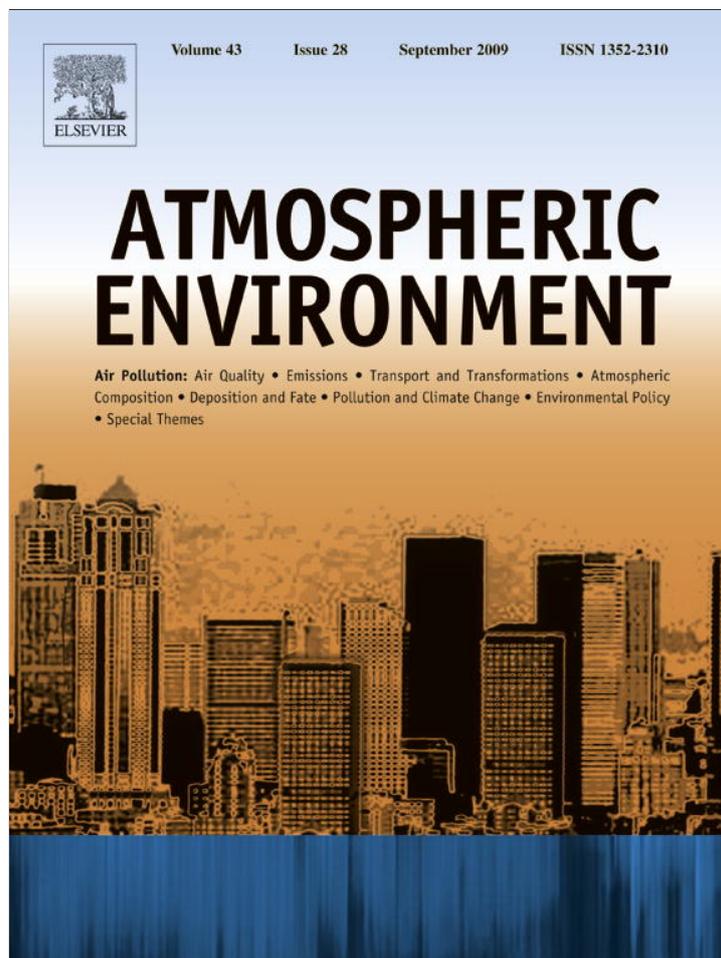


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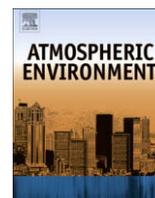
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The contribution of biological particles to observed particulate organic carbon at a remote high altitude site

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ABSTRACT

Although a significant fraction of atmospheric particulate mass is organic carbon, the sources of particulate organic carbon (POC) are not always apparent. One potential source of atmospheric POC is biological particles, such as bacteria, pollen, and fungal spores. Measurements of POC and biological particles, including bacteria, fungal spores, and pollen, were made as part of the Storm Peak Aerosol and Cloud Characterization Study in Steamboat Springs, CO in March–April 2008. Biological particles were identified and characterized using several methods. The results suggest that biological particles could account for an average of 40% of the organic carbon mass in particles with aerodynamic diameters less than 10 μm . These estimates of POC mass from biological particles are highly uncertain; however, the results suggest that biological particles could be a significant source of organic aerosol in the background continental atmosphere and further observations are needed to better constrain these estimates.

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1. Introduction

Atmospheric aerosols impact climate through direct and indirect forcing, degrade air quality and visibility, and have detrimental effects on human health, and are therefore important on many scales. Particulate organic carbon (POC) is a significant component of atmospheric aerosols in polluted urban airsheds as well as in more remote regions (e.g., Zhang et al., 2007). Despite the importance of POC in the atmosphere, the sources of POC are often undetermined. Many model simulations of POC in polluted atmospheres underestimate the POC mass by as much as an order of magnitude when compared to observations (e.g., Heald et al., 2005), whereas remote areas show closer agreement, but are nonetheless highly uncertain due to slow rates of POC formation (Tunved et al., 2006). Possible explanations for this model/measurement discrepancy include incorrect emission estimates of primary POC and precursor volatile organic compounds, missing precursors of secondary organic aerosol production, and missing

chemical and physical processes that lead to secondary organic aerosol production. Several studies suggest that a main source of measured POC is modern, in other words not derived from fossil fuel sources. For example, Ke et al. (2007) used organic tracer-based chemical mass balance (CMB) modeling and radiocarbon (C-14) measurements in the Tennessee Valley Region of the eastern U.S. and determined that as much as 84% of the observed summertime POC was contemporary (i.e., modern carbon, and not from fossil fuel combustion).

One major source of contemporary POC is secondary aerosol formation via the photo-oxidation of biogenic volatile organic compounds. Although this is estimated to be a significant source of atmospheric POC (e.g., Henze and Seinfeld, 2006), inclusion of secondary aerosols does not necessarily rectify the aforementioned model/measurement discrepancies. For example, Sakulyanontvittaya et al. (2008) show that inclusion of biogenic sesquiterpene emissions and subsequent secondary aerosol formation improves model performance in regional chemical model simulations of the U.S.; however, the model results still underpredict POC when compared to network observations. Another source of POC to the atmosphere that is not currently considered in most model simulations is primary biological particles, which include bacteria, fungal spores, and plant

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pollen. Recent studies suggest that primary biological particles can contribute significantly to atmospheric POC; however, the results of these studies are limited and vary substantially. Based on measurements in the Amazon, Elbert et al. (2007) attributed an average of 35% of the total aerosol mass to be fungal spores. At a continental background site in the Austrian Alps in March 2000, Bauer et al. (2002b) measured biological components in atmospheric samples and estimated that bacteria comprise to 0.03% and fungal spores 0.9% of the total observed POC mass. However, at a suburban site, Bauer et al. (2008) report that fungal spores comprise 6% and 14% of the POC mass concentration in the spring and summer, respectively. Not only are primary biological particles potentially an important contribution of organic aerosol in the atmosphere, specific microbial species may directly influence climatic conditions by acting as cloud and ice nucleators (e.g., Vali, 1971, 1996).

In March and April 2008, measurements of POC and biological particles were made at a remote mountain laboratory. Using these measurements, the potential contribution of biological particles to the observed mass of particulate organic carbon with diameters less than 10 μm (POC10) is estimated. The results of the observations and analysis suggest that the biological component of the observed POC10 was substantial and further study is warranted.

2. Methods

The Storm Peak Aerosol and Cloud Characterization Study (SPACCS08) was conducted from March 24 through April 15, 2008 at the Storm Peak Laboratory (SPL), located on top of Mt. Werner within the Steamboat Springs ski resort in Colorado (40.45°N, 106.73°W; 3200 m ASL). Measurements of meteorological variables, particle number and size distribution, organic and elemental carbon mass concentrations in particles with aerodynamic diameters less than 10 μm , and biological particle concentrations and composition were completed as part of SPACCS08. Site meteorology parameters, including wind speed and direction, temperature, relative humidity, and pressure, were observed throughout the study at 5 min intervals. Measurements of particle number concentrations and size distributions of particles with aerodynamic diameters from 530 nm to 18.4 μm were made with a TSI Aerodynamic Particle Sizer (TSI APS; Model 3320). The instrument was calibrated by the manufacturer (TSI) in October 2007. The instrument flow was checked previous to the SPACCS08 campaign. The number and size distribution of particles with aerodynamic diameters from 10 to 330 nm were measured with a TSI Scanning Mobility Particle Sizer (SMPS; Model 3396L22 with a TSI 3022 Condensation Particle Counter). Particles measured with the APS and SMPS were sampled at a flow rate of $\sim 50 \text{ L min}^{-1}$ from an insulated, 15 cm diameter manifold within approximately 1 m of its horizontal entry point through an outside wall. The 4 m high vertical section outside the building is capped with an inverted can to exclude cloud and ice. The APS, SMPS, and meteorological data are collected regularly as part of the SPL instrument suite.

As part of SPACCS08, a semi-continuous Sunset Laboratory Organic and Elemental Carbon (OC/EC) thermo-optical transmission analyzer (Birch and Cary, 1996) was deployed at SPL. This field instrument was compared with the laboratory-based NIOSH 5040 method (NIOSH, 1996; Bae et al., 2004) when both were run off the same inlet for an entire year as part of the St. Louis-Midwest Supersite. Excellent agreement was observed with a coefficient of regression, r^2 , for total carbon (EC + POC) of 0.89 and an r^2 for POC of 0.90. The precision of the field instrument was evaluated by as part of the Southern California Supersite (Arhami et al., 2006) as two instruments were run side-by-side. The r^2 for side-by-side hourly POC was 0.98 and 0.97 for hourly EC. For this study, atmospheric samples were collected at a flow rate of 6.2 L m^{-1} through

a URG cyclone, which restricted the sampling to those particles with diameters less than 10 μm . Incoming air samples were passed through a carbon paper organic gas denuder (provided by Sunset Labs) and collected on a quartz filter in the instrument. The standard four-step helium environment heating procedure (340, 500, 615, and 870 °C) was used to analyze POC. Samples were collected over a 4 h period to maximize the number of measurements with EC and OC above detection limits (manufacturer states detection limit of 0.05 $\mu\text{g m}^{-3}$ for a 4 h sample) while still providing a high time resolution image of aerosol properties. OC/EC measurements were made from March 31 through April 15, 2009.

Biological particles (bacteria, fungal spores, and plant pollen) were collected from a separate system comprised of two 0.2 μm cellulose nitrate filters (Fisher Scientific, Pittsburgh, PA). Ambient air was pulled through each of two filters simultaneously with a flow rate of 7.5 L min^{-1} per filter for periods of time that ranged from 5.5 to 12.25 h. Samples were collected at 9 different time periods from March 23 to April 07, 2008 and the total volume of air collected during each sampling period ranged from 2.5 m^3 to 5.4 m^3 per filter. After each sampling period, the two filters were immediately frozen at $-20 \text{ }^\circ\text{C}$. One filter of each set was used for total microbial abundance measurements via epifluorescence microscopy and the second used for DNA extraction and microbial community composition analyses. For the first, particles (including microbial cells) were shaken from the filters into 8 mL of HPLC-grade water in a small Petri dish for 2 h at 4 °C. A subset of the filters was examined under a microscope after this shaking process to assure that most visible particles were removed from the filter. While the efficiency of particle removal was not evaluated for every individual sample, all sample filters were treated identically, and therefore any bias associated with this method should be held constant across the sample set. The particles were stained with 4'-6'-Diamidino-2-phenylindole (DAPI), a DNA binding dye (KPL, Gaithersburg, MD) at a final working concentration of 500 $\mu\text{g mL}^{-1}$, and counted at 1000 \times magnification using a Nikon Eclipse E400-epifluorescence microscope following a similar protocol to the one described in Hernandez et al. (1999). Briefly, intact cells were counted on 25 mm diameter black polycarbonate filters with pore sizes of 0.22 μm (GEI-W&PT, Trevose, PA). Microbial abundance is expressed as cells per cubic meter of air, taking into account the dilution, the flow rate, and the number of hours sampled.

The second filter of each set was used to measure the relative abundance of bacteria, fungal spores, and pollen in the air samples. This was determined using the molecular, sequence-based technique described in Fierer et al. (2008). Briefly, the Ultra-Clean Plant DNA isolation kit (MoBio Laboratories, Carlsbad, CA) was used to extract the DNA from microbial cells trapped on the filters. Small-subunit rRNA genes were amplified using a universal PCR primer set (515F, 1391R; Angenent et al., 2005), the amplicons were cloned using the TOPO TA cloning kit (Invitrogen), and on average, 53 clones per sample were sequenced at Agencourt Bioscience (Beverly, MA). The sequences were then assigned to taxa using the BLAST algorithm against the GenBank nr database, assuming that the proportional representation of sequences in each clone library reflects the representation of bacterial, fungal, and plant pollen cells in each collected air sample. Considering the fact that DNA is very unstable in the free environment (e.g., when not contained within a cell) due to UV radiation and other atmospheric stresses, this method should allow us to assess the relative abundances of bacterial, fungal, and pollen cells in the atmosphere.

Five of the biological particle samples had corresponding POC10 measurements. Not all of the samples were coincident due to the timing of the different sampling techniques, and instrument malfunction of the OC/EC analyzer that required it to be offline for part of the study.

3. Results

Averaged over the study period from March 24 to April 15, 2008, hourly total particle number concentrations measured with the SMPS and the APS averaged 1900 cm^{-3} and 140 cm^{-3} , respectively (Fig. 1). These concentrations are similar to those reported by Lowenthal et al. (2002) during a study at SPL in February–May, 2001, where they observed particle number concentrations (particles $> 10 \text{ nm}$) of 1221 and 3893 cm^{-3} during cloudy and clear conditions, respectively. These values are typical of continental to remote regions, where particle number concentrations characteristically range from 2000 to $10,000 \text{ cm}^{-3}$ (Seinfeld and Pandis, 1998). The average number concentrations of the smaller particles measured by the SMPS showed a diurnal profile (Fig. 1a), which is suspected to be due to the mixing up of the boundary layer air from the valley below with clean tropospheric air and the resulting aerosol formation. These types of observations have been observed at other sites (e.g., Shaw, 2007). Although daytime increases in the small particle number concentrations were observed regularly, the

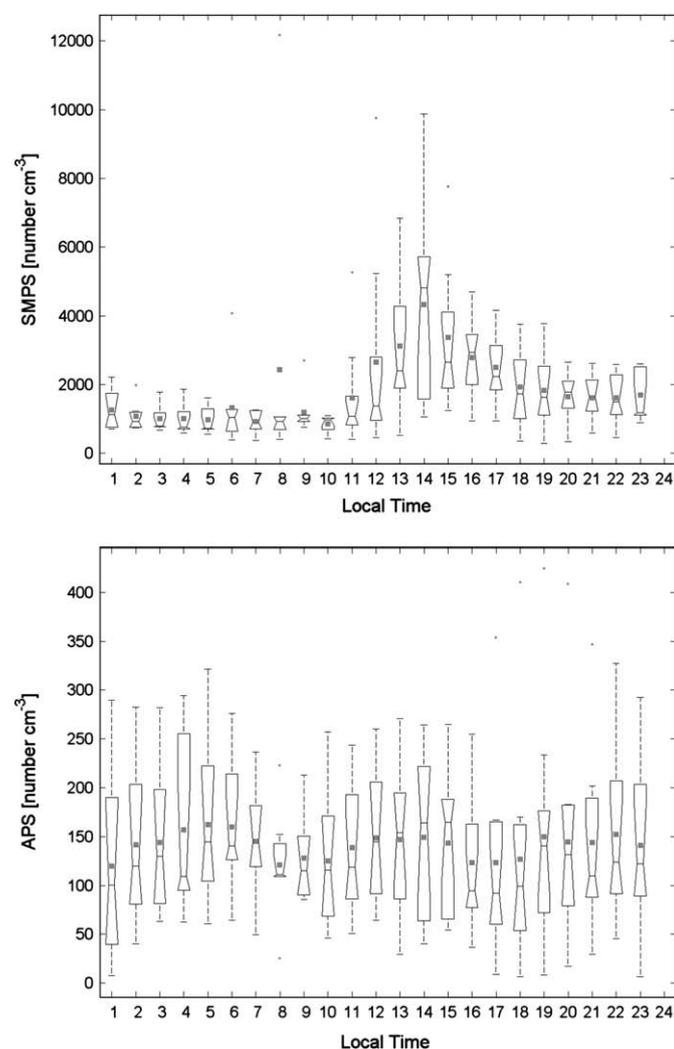


Fig. 1. Average diurnal particle number concentrations (March 30–April 10, 2008) of 10–330 nm and 0.5–5 μm and particles measured by the SMPS (1a, top) and APS (1b, bottom), respectively. Explanation of symbols in boxplot: Vertical lines represent the lower quartile, median, and upper quartile values. Means are shown as gray squares. Whiskers extend from each end of the box to the adjacent values in the data; the most extreme values within 1.5 times the interquartile range from the ends of the box. Outliers are data with values beyond the ends of the whiskers and are displayed with a gray dot.

larger particle ($>0.5 \mu\text{m}$) number concentrations do not appear to show this same diurnal trend (Fig. 1b). Since the larger particles comprise the majority of the measured particulate mass, we assume that the observed particulate mass was not significantly influenced by diurnal variations and was more indicative of the regional background aerosol that has experienced much longer atmospheric lifetimes.

Between April 01 and April 07, when the biological particle and POC10 measurements were coincident, the measured POC10 ranged from 0.39 to $1.64 \mu\text{g m}^{-3}$, with an average of $0.74 \mu\text{g m}^{-3}$ (Table 1). These observations show no distinct diurnal profile and do not appear to have been influenced by the meteorological conditions. The POC10 measurements are consistent with organic aerosol measurements made at SPL in April and May 2004, where an average of $\sim 1 \mu\text{g m}^{-3}$ organic aerosol (diameters $< 1 \mu\text{m}$) were observed (Zhang et al., 2007; E. Dunlea, *personal communications*). The observed POC concentrations are also similar to those observed at other remote high mountain sites. For example, measurements of POC at a high alpine site in the Swiss Alps (Jungfraujoch; 3580 m.a.s.l.), where a Sunset Labs OC/EC instrument was deployed between February and March 2005, were up to $\sim 2 \mu\text{g m}^{-3}$, although concentrations were typically $< 1 \mu\text{g m}^{-3}$ (Cozic et al., 2008). Based on a comparison with other observations from remote continental mountain sites, the observations made as part of SPACCS08 were very similar in magnitude, suggesting that this site is representative of remote, high altitude background concentrations.

Biological particle concentrations and community composition were determined by DAPI counts (Bauer et al., 2002b) and ribosomal DNA sequencing, respectively. The ribosomal sequencing approach allowed us to determine the approximate proportions of bacteria, fungi, and pollen for each of the nine air samples. Overall, plant pollen contributed the least to the DNA-containing particles (average of 4%): fungal spores and bacteria made up the majority (Table 2). DNA analysis of the biological particles shows a consistent species distribution through all samples, suggesting that the airborne microbial abundance and community composition of the high alpine environment is surprisingly stable throughout the shifting atmospheric conditions (Bowers et al., *in press*). This result is consistent with work performed by Despres et al. (2007), as the air above the high alpine environment was found to be the least microbially diverse of three surveyed landscapes (urban, rural, and high-alpine).

Using the results of the biological particle analysis and the measured concentrations of POC10 from SPL during SPACCS08, we attempt to constrain the relative contributions of biological particles to measured POC10 in remote, continental air masses. The number concentration of biological particles was on the order of 10^6 m^{-3} , ranging from 9.56×10^5 to $6.61 \times 10^6 \text{ m}^{-3}$ (Table 1). Because pollen is typically larger than $10 \mu\text{m}$ in diameter (e.g., Ingrouille and Eddie, 2006), it is assumed that none of the observed pollen was measured by the Sunset Labs OC/EC instrument. Additionally, based on a qualitative assessment during the DAPI counts, approximately 1–10% of the biological particles counted were larger than $10 \mu\text{m}$. To account for these larger particles that were identified in the biological particle analysis, but would not have been measured by the OC/EC instrument, we assume that only 90% of the total measured biological particle number concentrations represent particles observed by the OC/EC instrument.

To estimate the mass of the observed biological particles, an empirical fit was used to estimate the dry weight (DW) of the biological particles as a function of the cell volume (V): $DW = 485 \times V^{0.86}$ (Loferer-Krossbacher et al., 1998), assuming an average cell size to estimate the volume of the observed biological particles measured at SPL. Bacteria typically range in size from $0.5 \mu\text{m}$ to $2.5 \mu\text{m}$ (Wiedinmyer et al., 2006 and references therein)

Table 1

Measured number concentrations of total particles $>0.5 \mu\text{m}$, DAPI stained particles. Biological particle mass concentrations are calculated assuming a $1 \mu\text{m}$ diameter. POC10 is the average coincident measurement.

Sample ID	Date	Description	APS total particle counts ($\# \text{cm}^{-3}$)	Total DAPI stained ($\# \text{cm}^{-3}$)	90% DAPI stained ($\# \text{cm}^{-3}$)	Estimated DAPI mass ($\mu\text{g m}^{-3}$)	POC10 ($\mu\text{g m}^{-3}$)	Percent biological
3–23.CLN	3/23/08	Clear, Night	xxx	5.1	4.6	0.58 (0.1–3.5)		
3–24.CLD	3/24/08	Clear, Day	xxx	5.4	4.9	0.61 (0.1–3.6)		
4–1.CD.N	4/1/08	Cloudy, Night	87	1.5	1.4	0.17 (0.03–1.0)	1.06	16 (3–94)
4–2.CD.N	4/2/08	Cloudy, Night	126	4.7	4.2	0.53 (0.09–3.2)		
4–3.CD.D	4/3/08	Cloudy, Day	127	6.6	5.9	0.74 (0.12–4.4)		
4–3.CL.N	4/3/08	Clear, Night	227	5.7	5.1	0.64 (0.11–3.8)	1.02	62 (11–370)
4–4.CL.N	4/4/08	Clear, Night	148	2.2	2.0	0.24 (0.04–1.5)	0.69	35 (6–220)
4–5.CD.D	4/5/08	Cloudy, Day	199	1.0	0.9	0.11 (0.02–0.64)	0.63	17 (3–102)
4–6.CD.N	4/6/08	Cloudy, Night	82	3.1	2.8	0.35 (0.06–2.1)	0.54	64 (11–390)
					Average	0.44 (0.07–2.6)	0.79	40

and Bauer et al. (2002a,b) suggest that the average size of atmospheric fungal spores is $2 \mu\text{m}$. For this analysis, the observed biological particles are assumed to be spherical, with an average diameter of $1 \mu\text{m}$. Fifty percent of the calculated cell dry weight is assumed to be carbon (Bauer et al., 2002a, 2008). Using these assumptions and the number concentration of biological particles determined with the DAPI counts, the biological particle mass concentrations are estimated to range from 0.1 to $0.7 \mu\text{g m}^{-3}$ (for all sampling dates), with an average of $0.4 \mu\text{g m}^{-3}$. The result is highly sensitive to the assumptions made. For example, if an average biological diameter of $0.5 \mu\text{m}$ is assumed, the average mass concentration calculated from all samples is $0.07 \mu\text{g m}^{-3}$, and the average is $2.6 \mu\text{g m}^{-3}$ for an assumed diameter of $2 \mu\text{m}$. The range of POC10 during the time period corresponding to the biological particle measurements ranged from 0.32 to $1.64 \mu\text{g m}^{-3}$, with an average of $0.69 \mu\text{g m}^{-3}$. For the five coincident measurements, the estimated biological particle mass concentration (assuming a $1 \mu\text{m}$ diameter) contributed from 16 to 64% of the measured POC10, with an average of 40% (Table 1).

The results of this analysis suggest that the contribution of biological particles to the POC mass concentrations at SPL could be substantial. However, the results are highly uncertain. Because only a limited number of the measurements of POC10 and biological particles were coincident, there is added uncertainty associated with our attempt to correct for the different size cuts used for the two sampling techniques. Additionally, if many bacteria were deposited to the biological particle collection filters while attached to larger particles (e.g., dust), then this would lead to an overestimation of the number of biological particles smaller than $10 \mu\text{m}$. Assumed values of the carbon content of bacteria cells and fungal spores are variable and can produce varying estimates, as do the estimates of biological particle size. The measurements made

Table 2

Fractional contribution of bacteria, fungal spores, and plant pollen to the total biological particles. The fractional abundances were measured via ribosomal sequencing and microbial community characterization, while the total number of biological particles (90% DAPI stained) was estimated via DAPI staining and counting.

Counted sample ID	90% DAPI stained ($\# \text{cm}^{-3}$)	Total number of sequences	Bacteria	Fungi	Plant pollen
3–23.CLN	4.6	39	0.36	0.56	0.08
3–24.CLD	4.9	62	0.60	0.39	0.02
4–1.CD.N	1.4	79	0.19	0.76	0.05
4–2.CD.N	4.2	45	0.53	0.44	0.02
4–3.CD.D	5.9	51	0.53	0.47	0.00
4–3.CL.N	5.1	55	0.56	0.44	0.00
4–4.CL.N	2.0	63	0.60	0.25	0.14
4–5.CD.D	0.9	51	0.59	0.35	0.06
4–6.CD.N	2.8	54	0.65	0.35	0.00
Average	3.5	55	0.51	0.45	0.04

during this campaign were limited in number and were only taken during a short episode in the spring. These results may not be representative of other times during the year.

Despite the large uncertainties associated with the measurements and analysis, the results presented here suggest that biological components can potentially contribute significantly to atmospheric POC in remote, continental sites and that future study of this source is warranted. Even if the smallest diameter is assumed for the biological particles (500nm), the contribution of biological material to the observed POC10 is estimated to be 3–10%. To better constrain estimates of the biological particle contribution to POC mass in the atmosphere, coincident measurements of POC mass and biological particles are needed, as are better estimates of atmospheric biological particle size and carbon content. Further, genetic analysis of measured organisms is necessary to assess all atmospheric biological components. As highlighted by Pace (1997), culturable organisms represent only a small fraction of the biological organisms that are identified in the atmosphere. Until recently, biological particles were not considered to contribute significantly to atmospheric POC. One reason (among others) is that culture-based estimates of bacterial and fungal cell numbers usually underestimate cell numbers by many orders of magnitude since most microbial cells can not grow in culture. As more studies adopt direct cell counting techniques as done in this study, instead of relying on cultivation-based cell counts, the estimated significance of primary biological particles in the atmosphere is likely to increase.

4. Conclusions

The results of this study suggest that biological particles may significantly contribute to the mass of organic carbon observed in atmospheric aerosols. This confirms some results suggested by C^{14} measurements. The observations presented here imply that, in order to fully understand the sources of atmospheric POC, biological particles must be considered. This means that measurements of these components should be included in future field studies, and that model studies need to include the emissions and transport of biological particles in the atmosphere. Because of the lack of data, a complete emissions inventory of biological particles has not been completed. However, to accurately simulate POC, both for realistic representation of climate and air quality, this needs to be accomplished. The results of this study highlight the need to integrate biological sciences with atmospheric sciences.

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