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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.

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 though 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 manufacture (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 ~50 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 SPACC08, 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 μg 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 °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 μg mL^{-1}, and counted at 1000× 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.