RESEARCH ARTICLE

Key Points:
- Black carbon aerosols primarily had a fossil fuel origin, while OC had a significant biogenic and biomass burning fraction that was higher in summer
- The sources of BC and OC observed in Salt Lake City were consistent with measurements from other large metropolitan areas outside the U.S.
- Using radiocarbon to monitor BC and OC aerosol sources may provide information about the success of different mitigation policies

Supporting Information:
- Supporting Information S1

Correspondence to:
G. O. Mouteva,
gmouteva@uci.edu

Using radiocarbon to constrain black and organic carbon aerosol sources in Salt Lake City

Gergana O. Mouteva[1] [iD], James T. Randerson[1] [iD], Simon M. Fahrni[1,2], Susan E. Bush[3], James R. Ehleringer[3] [iD], Xiaomei Xu[1], Guaciara M. Santos[1] [iD], Roman Kuprov[4], Bret A. Schichtel[5] [iD], and Claudia I. Czimczik[1]

[1]Department of Earth System Science, University of California, Irvine, California, USA, [2]Now at the Eidgenössische Technische Hochschule (ETH), Zürich, Switzerland, [3]Department of Biology, University of Utah, Salt Lake City, Utah, USA, [4]Division of Air Quality, Utah Department of Environmental Quality, Salt Lake City, Utah, USA, [5]National Park Service, Washington, District of Columbia, USA

Abstract Black carbon (BC) and organic carbon (OC) aerosols are important components of fine particulate matter ($PM_{2.5}$) in polluted urban environments. Quantifying the contribution of fossil fuel and biomass combustion to BC and OC concentrations is critical for developing and validating effective air quality control measures and climate change mitigation policy. We used radiocarbon (^{14}C) to measure fossil and contemporary biomass contributions to BC and OC at three locations in Salt Lake City, Utah, USA, during 2012–2014, including during winter inversion events. Aerosol filters were analyzed with the Swiss_4S thermal-optical protocol to isolate BC. We measured fraction modern (f_M) of BC and total carbon in $PM_{2.5}$ with accelerator mass spectrometry and derived the f_M of OC using isotope mass balance. Combined with ^{14}C information of end-member composition, our data set of 31 ^{14}C aerosol measurements provided a baseline of the fossil and contemporary biomass components of carbonaceous aerosol. We show that fossil fuels were the dominant source of carbonaceous aerosol during winter, contributing 88% (80–98%) of BC and 58% (48–69%) of OC. While the concentration of both BC and OC increased during inversion events, the relative source contributions did not change. The sources of BC also did not vary throughout the year, while OC had a considerably higher contemporary biomass component in summer at 62% (49–76%) and was more variable. Our results suggest that in order to reduce $PM_{2.5}$ levels in Salt Lake City to meet national standards, a more stringent policy targeting mobile fossil fuel sources may be necessary.

1. Introduction

Anthropogenic emissions of carbonaceous aerosol from fossil fuel and biomass combustion have increased significantly since 1850 [*Novakov*, 2003; *Bond et al.*, 2007; *Bauer et al.*, 2010] and account for a major fraction of the fine particulate matter ($PM_{2.5}$ aerodynamic mass diameter < 2.5 μm) in the global atmosphere, particularly in polluted metropolitan areas. The carbonaceous aerosol is often separated into two components: black carbon (BC) and organic carbon (OC) [*Penner and Novakov*, 1996; *Gelencser*, 2004]. Black carbon is emitted as a primary product of incomplete combustion of fossil fuels (e.g., coal, oil, natural gas) and contemporary biofuels (e.g., wood, crop residues) and thus often has an anthropogenic origin [*Haywood and Boucher*, 2000]. Urban BC loads and properties therefore typically reflect regional fuel consumption patterns such as fuel type and combustion technology [*Szidat et al.*, 2007; *Andersson et al.*, 2015]. In contrast, OC can be formed either as a primary aerosol from incomplete combustion of biomass or fossil fuels or by means of oxidation and condensation of volatile organic compounds (VOCs) emitted from biogenic or anthropogenic sources [*Griffin et al.*, 1999; *Kanakidou et al.*, 2005; *Goldstein and Galbally*, 2007]. The production of BC and OC from fuel burning depends on fuel type and combustion efficiency. Most biomass burning emissions of BC and OC originate from landscape fires in tropical savannas, forests, and peatlands [*Van der Werf et al.*, 2010], although other important sources include emissions from residential combustion and agricultural waste burning in South Asia [*Gustafsson et al.*, 2009; *Budhavant et al.*, 2015].

As a major component of $PM_{2.5}$, BC and OC contribute to the adverse health effects associated with $PM_{2.5}$ exposure. $PM_{2.5}$ has been linked to both respiratory [*Sheppard et al.*, 1999; *Knaapen et al.*, 2004; *Götschi et al.*, 2008; *Pope et al.*, 2011; *Young et al.*, 2014; *Rice et al.*, 2015] and cardiovascular [*Dominici et al.*, 2006; *Miller et al.*, 2007; *Brook et al.*, 2010; *Pope et al.*, 2011; *Kloog et al.*, 2015] diseases and, ultimately, increases

in morbidity and mortality [*Dockery and Pope*, 1994; *Schwartz et al.*, 1996; *Ostro et al.*, 2006; *Fann et al.*, 2012; *Johnston et al.*, 2012]. PM$_{2.5}$ also contributes to neurological deficits influencing intelligence quotient and cognition [*Oudin et al.*, 2016; *Chen et al.*, 2017]. Outdoor air pollution, mostly in the form of PM$_{2.5}$, was estimated to have caused 3.3 million premature deaths worldwide in 2010, with model projections indicating that these impacts could double by 2050 [*Lelieveld et al.*, 2015].

Urban air pollution can also contribute to the urban heat island effect [*Cao et al.*, 2016] and affect regional precipitation patterns [*Rosenfeld*, 2000]. In the physical climate system, BC and OC play important and often contrasting roles, as they influence climate directly through changes in absorption, scattering, or reflection of solar radiation [*Penner*, 1995; *Myhre et al.*, 2013] and indirectly through cloud microphysical interactions [*Ackerman et al.*, 2000; *Cozic et al.*, 2007; *Koch and Del Genio*, 2010]. Black carbon deposition on snow and ice further contributes to a positive climate feedback in mountain and high-latitude regions by decreasing snow albedo and accelerating melt rates [*Wiscombe and Warren*, 1980; *Flanner et al.*, 2007; *Jiao et al.*, 2014].

Within developed countries, metropolitan areas are prime targets for aerosol emission reductions because of co-occurring health and climate benefits [*Grieshop et al.*, 2009; *Lelieveld et al.*, 2015; *West et al.*, 2016]. To develop effective air quality legislation, however, more information is needed about the contribution of different sources that are often difficult to disentangle in complex urban environments. Important discrepancies remain between top-down BC and OC source estimates obtained from receptor modeling [*Stone et al.*, 2007; *Chen et al.*, 2013] and bottom-up emission inventories derived from fuel consumption data and emission factors [*Bond et al.*, 2004, 2007; *Junker and Liousse*, 2006; *Bauer et al.*, 2010]. These uncertainties also limit our ability to develop realistic emission time series as boundary conditions for Earth system models and thus contribute to uncertainties in projections of future climate change [*Penner et al.*, 2010; *Vignati et al.*, 2010]. Top-down and bottom-up discrepancies are significantly larger for areas where emission monitoring and reporting is limited [*Winiger et al.*, 2017], as compared to regions where technology-based emission inventories are more advanced [*Winiger et al.*, 2016]. These regional differences highlight the importance of improving emission distribution and source attribution with the aim of developing reliable climate model predictions and effective mitigation strategies.

One approach for improving attribution of fossil fuel and contemporary biomass contributions to BC and OC is to measure the aerosol radiocarbon (^{14}C) signature [*Heal*, 2014]. Radiocarbon is a particularly effective tracer for separating emissions from fossil and biomass combustion because fossil emissions are ^{14}C free and biomass emissions have a characteristic ^{14}C/^{12}C ratio that is proportional to atmospheric carbon dioxide (CO_2) at the time of carbon fixation [*Reddy et al.*, 2002]. This source apportionment approach has been used to separate fossil and biomass sources in urban [*Gustafsson et al.*, 2009; *Szidat et al.*, 2009] and remote atmospheric environments [*Slater et al.*, 2002; *Gelencsér et al.*, 2007; *Barrett et al.*, 2015; *Mouteva et al.*, 2015b]. Widespread use of this approach has been limited in the past by technical challenges related to isolation of CO_2 from BC and OC aerosol fractions and by small sample sizes that limit the accuracy of accelerator mass spectrometry measurements [*Szidat et al.*, 2013].

Here we used a recently developed method for measuring ^{14}C in microscale aerosol samples [*Zhang et al.*, 2012; *Mouteva et al.*, 2015a] to quantify the temporal and spatial variation in fossil and contemporary biomass contributions to BC and OC in a major metropolitan region, Salt Lake City, Utah, USA. An improved understanding of fossil and contemporary biomass contributions to carbonaceous aerosols is of great importance in the Salt Lake Valley, because particulate air pollution in winter is considerable and is exacerbated during inversion events [*Silcox et al.*, 2012; *Whiteman et al.*, 2014]. High PM$_{2.5}$ concentrations within this metropolitan area have been linked to an increased number of asthma attacks and emergency hospital visits [*Beard et al.*, 2012]. The relative contributions of motor vehicles versus residential wood burning emissions to winter air pollution has been the subject of debate. While a wood burning ban has been implemented as a necessary air pollution control measure during inversion events [*Utah Division of Air Quality*, 2015], the Salt Lake Valley continues to experience nonattainment PM$_{2.5}$ episodes. Thus, major uncertainties remain with respect to the relative importance of the different combustion sources. Here our ^{14}C measurements allowed us to identify how fossil and contemporary biomass contributions varied seasonally and during inversion events over a 16 month period. Our study may provide additional data necessary for designing effective mitigation policies.
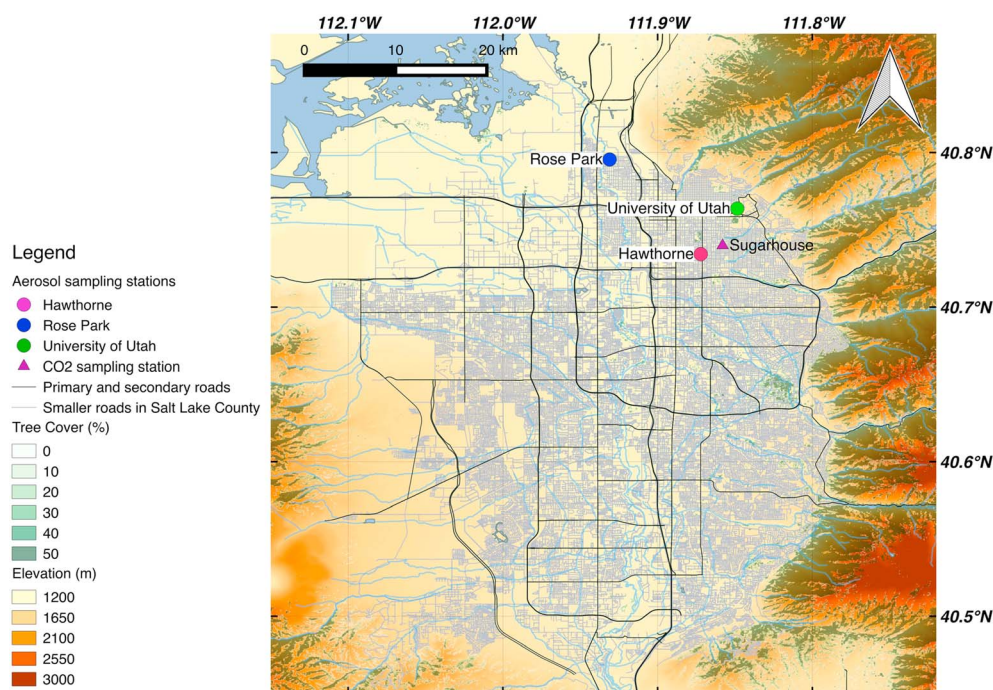
Figure 1. Study area. A map showing the location of the aerosol sampling stations within the Salt Lake City metropolitan area. The regional topography is from the Shuttle Radar Topography Mission 90 m Digital Elevation Data, percent tree cover is from the Hansen/UMD/Google/USGS/NASA forest cover data set [*Hansen et al.*, 2013], and road system and water layers are from the Topologically Integrated Geographic Encoding and Referencing (TIGER) data set, available at http://www.census.gov/geo/maps-data/data/tiger-line.html.

2. Experimental Methods

2.1. Sample Collection

We collected $PM_{2.5}$ at three locations across Salt Lake City (Figure 1, Table 1). The stations were selected to capture spatial variations within urban (Hawthorne), industrial (Rose Park), and suburban (University of Utah) areas of Salt Lake City. To capture seasonal variations, we collected samples over a 2 week period during each 3 month meteorological season from October 2012 to August 2013. Additional samples, targeting inversion periods, were collected at the Hawthorne station through the winter of 2013/2014. A total of 41 aerosol samples were collected: 26 at Hawthorne, 8 at Rose Park, and 7 at the University of Utah (Table A1). During our study period in 2013, the Salt Lake County population was 1.08 million [*Census Bureau and P. D.*, 2017].

At Hawthorne and Rose Park, samplers were positioned on Utah Division of Air Quality (DAQ) air-monitoring platforms elevated by 3 m or more above ground level. At the University of Utah, the sampler was installed on the roof of the Aline Skaggs Biology Building. Samples were collected on 20 cm × 25 cm quartz microfiber filters (2500 QAT-UP, Pallflex Tissuquartz, Pall, Port Washington, NY, USA), using high-volume total suspended particulate matter samplers (HIVOL-AMCLD, Thermo Environmental Instruments, Franklin, MA, USA) with SA-230-F impactor plates (TE-230-QZ, Tisch Environmental, Cleves, OH, USA), operated at 1130 L min^{-1}. A slotted

Table 1. Location of the $PM_{2.5}$ Sampling Stations[a]

Station	Latitude °N	Longitude °W	Elevation (m asl)	Site Description	EPA AIRS Code
Hawthorne[b]	40.7344	−111.8721	1306	Located in a metropolitan area in downtown Salt Lake City	490353006
Rose Park	40.7956	−111.9310	1295	Located in an industrial area in north Salt Lake City	490353010
University of Utah	40.7637	−111.8485	1426	A higher elevation site, located near the base of the Wasatch Mountains	N.A.

[a]The $PM_{2.5}$ sampling stations were colocated with the CO_2 monitoring network stations, except at Hawthorne. N.A. denotes not applicable.
[b]The closest CO_2 monitoring station at Hawthorne was the Sugarhouse CO_2 station, located at (40.740°N, 111.858°W, 1339 m asl).

microquartz fiber filter (TE-230-QZ, microquarts slotted collection substrates, Tisch) was installed on the impactor plate per sampling period to remove larger particles. The impactor filters were only used for sizing and were discarded following sampling. The sampling duration was set to approximately 48 h to allow accumulation of sufficient BC aerosol for ^{14}C analysis. A blank filter (one fourth the size of a sample filter) was mounted on the inside of each aerosol sampler housing (in an area without gas flow) during each sampling period to estimate the background contribution from volatile OC absorption (hereafter referred to as the blank).

Prior to sample collection, sample filters were prebaked at 500°C for 4 h, wrapped in aluminum foil, and stored in sealed plastic bags to prevent possible carbon contamination. Following sampling, the sample and blank filters were wrapped in aluminum foil, packed into the airtight plastic bags, and stored at −20°C.

2.2. Extraction of BC From Bulk PM$_{2.5}$

We used a thermal-optical OC/EC analyzer (Sunset Laboratory Inc., Tigard, OR, USA) with the Swiss_4S protocol [*Zhang et al.*, 2012], specifically developed for accurate ^{14}C analysis. When isolated with such thermal-optical approaches, the BC fraction is also commonly referred to as elemental carbon (EC) [*Bond et al.*, 2013]. The Swiss_4S method was adapted at the University of California Irvine, where a simple and efficient vacuum line was coupled to the OC/EC analyzer, allowing cryogenic trapping of the evolving CO_2 during the thermal-optical analysis with high accuracy and a low carbon blank [*Mouteva et al.*, 2015a]. The PM$_{2.5}$ filter samples were introduced into the OC/EC analyzer as 1.0×1.5 cm^2 filter punches (1.0 cm × 1.5 cm). Prior to analysis, punches were washed with Milli-Q water (Synergy 185, EMD Millipore, Billerica, MA, USA) and dried at 60°C for up to 1 h to remove water-soluble OC and minimize charring. To avoid bias in the ^{14}C measurement of BC from charring and premature BC loss, the Swiss_4S method discards a middle fraction as potential mixture of OC and BC and consequently may underestimate BC concentration [*Zhang et al.*, 2012] (see supporting information).

2.3. Measurement of the Total Carbon in Bulk PM$_{2.5}$

We separately analyzed the total carbon (TC) content of each PM$_{2.5}$ sample by taking multiple punches (n = 2–18, based on carbon content) from each filter. A conventional "sealed-tube" procedure was used to convert TC into CO_2. Punches were sealed with a known amount of cupric oxide under vacuum in prebaked 9 mm outer diameter quartz tubes and combusted at 900°C for 3 h. We then measured the mass of the TC-derived CO_2 and collected the CO_2 in a reaction tube for subsequent graphitization and ^{14}C analysis [*Xu et al.*, 2007]. The OC concentrations were calculated as the difference between TC and BC.

2.4. ^{14}C Analysis of Carbonaceous Aerosols

The ^{14}C content of BC and TC was measured with accelerator mass spectrometry at the W. M. Keck Carbon Cycle Accelerator Mass Spectrometry laboratory (KCCAMS) at the University of California Irvine. The OC ^{14}C content was estimated using isotope mass balance equations. Prior to analysis the purified CO_2 was converted into a solid graphite target. Different methods were used for graphitization of the BC and TC fractions due to their different carbon masses (calculated as mg C). The smaller BC samples ($<$ 0.01 mgC) were graphitized using an open-tube hydrogen reduction method specifically optimized for ultrasmall samples [*Santos et al.*, 2007]. The larger TC samples ($>$ 1 mg C) were graphitized using a sealed-tube zinc reduction method [*Xu et al.*, 2007]. All samples were processed alongside standards and blanks [*Xu et al.*, 2007; *Mouteva et al.*, 2015a]. The ^{14}C data is reported here as fraction modern (f_M), which represents the deviation of the ^{14}C/^{12}C ratio of the sample from modern, defined as 95% of the ^{14}C concentration of the standard OX-I (or NIST HOxI SRM 4990B) in 1950 [*Stuiver and Polach*, 1977], corrected for mass-dependent fractionation obtained from the online measurement of ^{13}C/^{12}C in the spectrometer [*Santos et al.*, 2007]. All of the individual sample carbon concentrations and f_M values, along with the date and duration of each sampling interval, are reported in Table A1. All 41 of the aerosol samples we collected had sufficient carbon mass to enable ^{14}C analysis of TC, and 31 had sufficient mass to enable ^{14}C analysis of BC.

2.5. Radiocarbon-Based Source Apportionment

A simple isotopic mass balance equation was applied using fraction modern signatures of fossil fuel ($f_{M(fossil)}$) and biomass ($f_{M(bio)}$) and the measured fraction modern signature of the BC or OC ($f_{M(C)}$), to calculate the

fossil-derived BC and OC (C_{fossil}) from the total concentration (denoted as C). We applied the same equation separately for BC and OC:

$$f_{M(C)} \times C = f_{M(fossil)} \times C_{fossil} + f_{M(bio)} \times (C - C_{fossil})$$

Fossil material is ^{14}C-free and therefore characterized by $f_{M(fossil)} = 0$, while contemporary material is intrinsically labeled with a characteristic ^{14}C/^{12}C ratio associated with the time of CO_2 fixation and the local atmospheric ^{14}C concentration. Biomass produced after 1955 is enriched in [14]C ($f_M > 1$) because aboveground thermonuclear weapons testing in the late 1950s and early 1960s produced large quantities of ^{14}C, causing a sudden twofold increase in the ^{14}C/^{12}C ratio of the Northern Hemisphere atmospheric CO_2. Since the partial cessation of nuclear testing in 1963, the global ^{14}C/^{12}C-CO_2 has been declining as a consequence of exchange with ocean and land carbon reservoirs and dilution by fossil fuel emissions [*Suess*, 1955; *Levin et al.*, 2010].

To estimate the contemporary biomass f_M end-member, we used direct ^{14}C measurements of two potential sources: (1) leaves, as a proxy for emissions of biogenic VOCs by plants or green waste burning, and (2) firewood smoke as a proxy for emissions from residential wood burning (Table 2). During summer, the OC fraction can have a large contribution from the oxidation of biogenic VOCs, which are often produced by means of different metabolic pathways in leaves and are closely coupled with carbon fixation pathways [*Kesselmeier and Staudt*, 1999; *Lerdau et al.*, 2013]. To capture the isotopic signature of the volatile organic source contribution, we analyzed mature live aspen leaves, collected in the Salt Lake Valley between June and August 2013.

Wood smoke can also be an important contributor to $PM_{2.5}$ throughout the year. To accurately represent this in our source apportionment analysis, we measured the ^{14}C content of TC in $PM_{2.5}$ from the combustion of three types of firewood purchased in Salt Lake City in the winter of 2013/2014. A wood burning stove in a laboratory setting at the University of Utah was used to collect the $PM_{2.5}$ samples on baked TE-230-QZ filters in March 2014.

In the source apportionment calculations, average values were used for all leaf samples ($f_{M(bio/leaves)}$ = 1.019 ± 0.002, $n = 14$) and all wood samples ($f_{M(bio/wood)}$ = 1.041 ± 0.035, $n = 4$) (Table 2). The greater fraction modern for the wood samples was consistent with the bole of trees accumulating carbon over a period of many prior years, at times when atmospheric ^{14}C/^{12}C ratios were greater [*Levin et al.*, 2010; *Mouteva et al.*, 2015b].

2.6. End-Member Analysis of Fossil and Contemporary Biomass Contributions

To more quantitatively estimate the relative contributions of fossil and contemporary biomass sources to BC and OC and to integrate important sources of uncertainty, we performed a two-source end-member analysis. For the contemporary biomass end-member, we developed two scenarios for the spring-summer-fall period using the mean of either the aspen leaf or wood f_M measurements as the end-member (Table 3). This approach recognizes that both oxidation of biogenic VOCs and combustion of biomass fuels likely contributed to the carbonaceous aerosol during these seasons.

For the winter, we used only the wood smoke samples for the contemporary biomass end-member, recognizing that production of biogenic VOCs was likely to be minimal at cold temperatures [*Guenther et al.*, 2006]. For f_M measurements of BC near the ^{14}C detection limit for ultrasmall graphite targets, we developed two additional scenarios to help quantify uncertainty. In one scenario, we assumed that the BC f_M upper limit was the correct aerosol measurement value. In the second scenario, we assumed that these samples were at the lower detection limit and contained no [14]C (i.e., f_M was equal to 0).

2.7. Additional Data Sources

Continuous observations of near-surface CO_2 mole fractions were obtained from the University of Utah's CO_2 measurement network (http://air.utah.edu). We report midafternoon mole fraction measurements of CO_2 (measured halfway between solar noon and sunset) from the following sites: Rose Park (40.794°N, 111.932°W, 1299 m above sea level (asl)), Sugarhouse (40.740°N, 111.858°W, 1339 m asl), and University of Utah (40.763°N, 111.848°W, 1430 m asl). We selected midafternoon measurements to better identify winter inversion events that often persisted for several days and contributed to elevated CO_2 that far exceeded background levels. Two of the CO_2 measurement sites were colocated with the aerosol sampling sites (Figure 1). The Sugarhouse CO_2 monitoring station was selected because it was the closest available

Table 2. Radiocarbon Composition of Samples Used to Estimate the Biomass End-Member

Description of Samples	n	Fraction Modern
Aspen leaves		
1.4 km radius of Hawthorne	5	1.018 ± 0.006
0.3 km radius of Rose Park	4	1.021 ± 0.003
0.4 km radius of University of Utah	5	1.020 ± 0.004
Average ± SD		1.019 ± 0.002
Wood		
Pine	2	1.078 ± 0.028
Oak	1	1.035 ± 0.002
Maple	1	1.009 ± 0.002
Average ± SD		1.041 ± 0.035

station to the Hawthorn aerosol sampling site. We also obtained daily 24 h-averaged $PM_{2.5}$ concentration for the Hawthorne monitoring station (https://www.epa.gov/outdoor-air-quality-data). The $PM_{2.5}$ was measured gravimetrically on 48 mm diameter teflon filters with a sequential air sampler for 24 h (Partisol 2025i, Thermo). We used these data to define inversion periods when the $PM_{2.5}$ concentrations exceeded the 24 h-average U.S. National Ambient Air Quality Standard (NAAQS) of 35 μg/m^3 continuously for more than 3 days. Since inversion periods are characterized by high atmospheric stability, synoptic-scale high-pressure systems, and persistent cold-air pools [*Gillies et al.*, 2010; *Silcox et al.*, 2012], we also used DAQ measurements of ambient temperature and barometric pressure, collected every day, to help interpret the seasonal dynamics of our ^{14}C observations.

3. Results

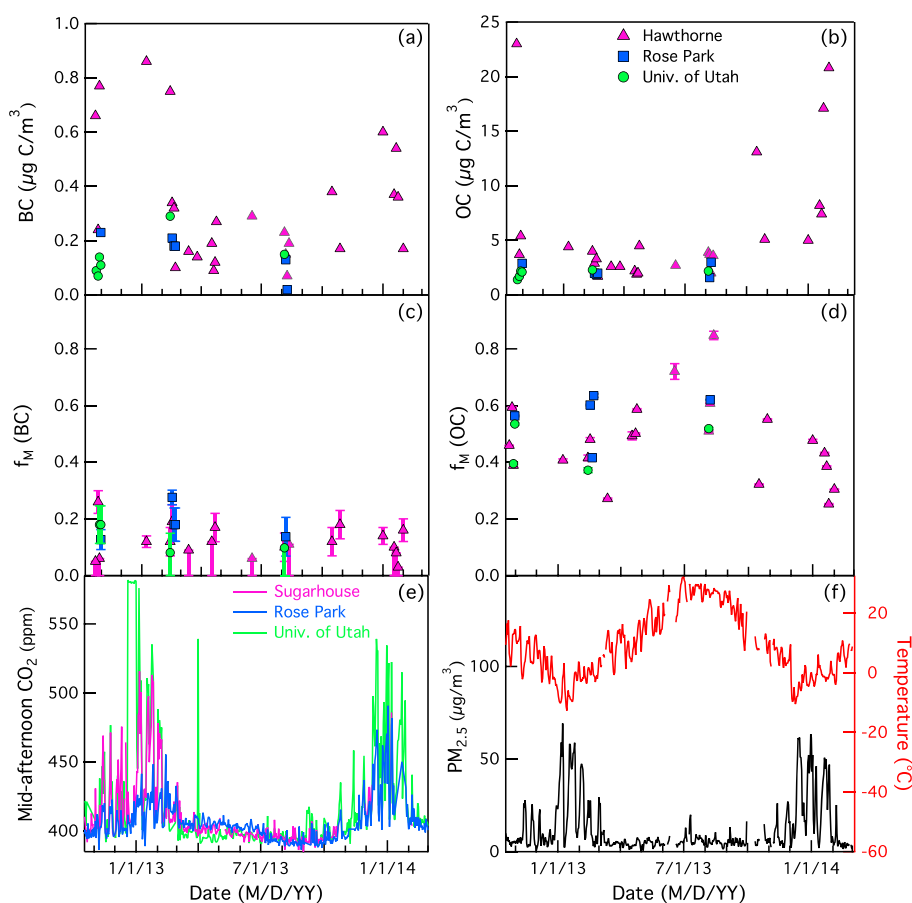### 3.1. Aerosol Concentration and Fraction Modern Observations

We observed the highest concentrations of BC and OC during winter and fall, which coincided with periods of elevated CO_2 and $PM_{2.5}$ (Figure 2). Elevated CO_2 is a typical characteristic of inversion events in Salt Lake City, where emissions of trace gases and aerosols are trapped in cold surface layers that inhibit convection and increase natural gas heating demands [*Pataki et al.*, 2003]. Black carbon concentrations were highest in winter, with a mean (± 1 standard deviation) of 0.38 ± 0.23 μg C/m^3 air ($n = 14$ samples), and lowest in summer with a mean of 0.15 ± 0.09 μg C/m^3 ($n = 8$). Winter and summer differences in BC concentrations were statistically significant at a 95% confidence level (analysis of variance) (Figure 3). In contrast, OC aerosols were more variable, particularly during fall and winter, and thus seasonal variations were not statistically different (Figures 2b and 3b). The mean OC for winter was 5.9 ± 5.9 μg C/m^3 ($n = 14$), and the mean for fall was 6.1 ± 6.9 μg C/m^3 ($n = 10$). In the spring and summer we observed lower OC concentrations and less variability, with a spring mean of 2.6 ± 0.9 μg C/m^3 ($n = 6$) and a summer mean of 2.8 ± 0.9 μg C/m^3 ($n = 8$).

Seasonal patterns of f_M were considerably different for BC and OC, indicating that the two aerosol components had different sources. The f_M of BC was relatively uniform temporally and spatially with an annual mean and standard deviation of 0.13 ± 0.11 ($n = 30$, Figures 2c and 3c). Summer and winter f_M levels were statistically indistinguishable. Several BC samples had very low $^{14}C/^{12}C$ ratios that were near or at the measurement detection limit (Table A1). For these samples, the reported f_M value shown in Figure 2c represents the most positive upper bound, and the reported measurement uncertainty range varied between 0 and this upper limit.

In contrast, the f_M of OC was considerably higher than that of BC and varied significantly between seasons (Figures 2d and 3d). The f_M of OC had an annual mean of 0.50 ± 0.24 ($n = 18$), with values during the winter (0.43 ± 0.11, $n = 12$) significantly lower than those during the summer (0.64 ± 0.13, $n = 6$) (two-sample t test). We did not observe any significant difference in concentration or f_M across the different stations, for either BC or OC. Therefore, in the following section we present results, averaged across all sites.

3.2. Radiocarbon-Derived Fossil and Biomass Source Contributions to BC and OC

Our analysis indicated that fossil fuels were a dominant contributor to BC throughout the year, with an annual mean of 89%, a range of 78 to 100%, and with no statistically significant differences among seasons (Table 3). In contrast, OC had a considerable non-fossil source contribution that varied seasonally. Non-fossil OC accounted for 62% (with a range of 49–76%) of the OC mass during summer and for 42% (31–52%) during winter. During summer, the higher non-fossil fraction of OC would be consistent with a larger contribution from the oxidation of biogenic VOCs. During winter, the end-member analysis was consistent with residential wood burning contributing significantly to OC mass but having a minimal influence on BC. The radiocarbon

Figure 2. BC and OC concentrations and fraction modern (f_M) results from three stations in Salt Lake City. Top row shows concentrations of (a) BC and (b) OC; middle row shows fraction modern values of (c) BC and (d) OC; and bottom row shows environmental variables from other sources during the period of our campaign. (e) Atmospheric CO_2 mole fractions were measured by the University of Utah network, and (f) 24 h-averaged $PM_{2.5}$ concentrations and ambient air temperature were measured by the Utah DAQ at the Hawthorn station.

source apportionment method, however, cannot distinguish between contemporary biomass burning and biogenic VOC contribution to the OC aerosol. To quantitatively evaluate the biogenic VOC contribution and distinguish it from biomass burning emissions, further analysis of other chemical tracers is required.

3.3. Winter Inversion Measurements of BC and OC

During winter inversions in the Salt Lake Valley, persistent cold-air pools and multiday episodes of high ambient atmospheric stability lead to exacerbated $PM_{2.5}$ levels [*Silcox et al.*, 2012]. Therefore, during the campaign we specifically aimed to investigate the sources of BC and OC during such conditions. We sampled two winter seasons (2012/2013 and 2013/2014), each characterized by three inversion events. Of the 41 samples collected in this study, we captured four winter inversion samples—one during the first winter and three during the second winter (Table A1). The isotopic composition of BC and OC during inversions was similar to their composition during non-inversion winter periods. The fraction modern means of BC and OC were statistically indistinguishable between inversion and non-inversion samples, and from our end-member analysis we found that on average 92% (range: 81–100%) of the BC and 59% (range: 55–63%) of the OC had a fossil origin during inversions.

4. Discussion

4.1. Implications for Effective Air Quality Policy in Salt Lake City

During winter inversion events, Salt Lake County often experiences episodes with $PM_{2.5}$ concentrations exceeding the 24 h-average NAAQS of 35 $\mu g/m^3$. Globally, this standard is even lower, as set by the
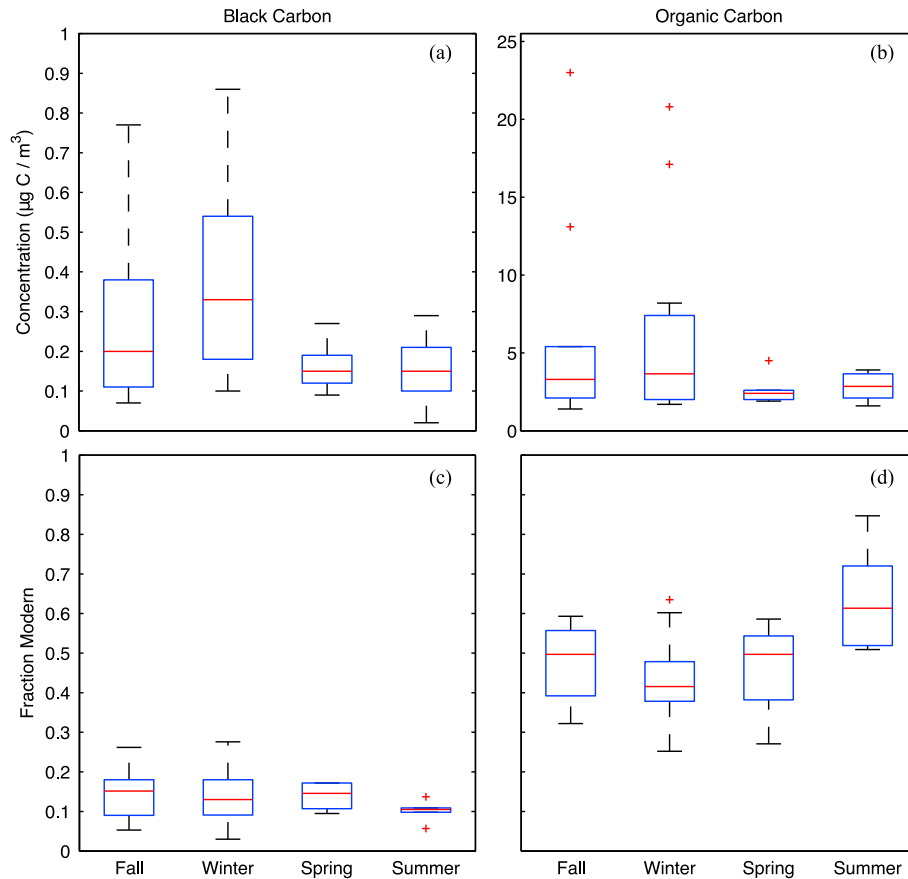
Figure 3. Box and whiskers plot of concentration and fraction modern (f_M) for BC and OC aerosol. Top row shows variation in (a) BC and (b) OC concentrations, and bottom row shows variation in the f_M of (c) BC and (d) OC for each season, combining observations from all stations together. For each box, the central red line is the median, the top and bottom edges of the box are the 75th and 25th percentiles, the whiskers extend to the most extreme data points not considered outliers, and outlier points are plotted individually.

World Health Organization (WHO) at 25 μg/m³. Despite continuous efforts to reduce emissions through the development of a comprehensive state implementation plan and ensuing regulations, Salt Lake County has continued to experience nonattainment days during winter inversion events [*Whiteman et al.*, 2014; *Utah Division of Air Quality*, 2015]. The nonattainment status of an area has legislative implication for air quality mitigation. Salt Lake City failed to meet the 24-h NAAQS for $PM_{2.5}$ by the Environmental

Table 3. Fossil Contribution (%) to BC and OC Aerosol Concentrations[a]

End-Member Assumptions	Fall		Winter[b]		Winter Inversion		Spring		Summer	
	BC	OC	BC	OC	BC	OC	BC	OC	BC	OC
					Wood combustion					
Upper limit f_M(BC)	86 ± 7	54 ± 10	87 ± 6	59 ± 10	89 ± 3	59 ± 4	87 ± 4	56 ± 13	90 ± 3	39 ± 12
At the limit f_M(BC) = 0	88 ± 9	54 ± 9	89 ± 9	58 ± 10	94 ± 7	59 ± 4	92 ± 10	55 ± 13	94 ± 6	38 ± 12
					Fresh Biomass					
Upper limit f_M(BC)	86 ± 7	53 ± 10	n.a.	n.a.	n.a.	n.a.	86 ± 4	55 ± 13	90 ± 3	38 ± 13
At the limit f_M(BC) = 0	87 ± 9	53 ± 10	n.a.	n.a.	n.a.	n.a.	92 ± 10	54 ± 13	94 ± 6	37 ± 13
Average	87	54	88	58	92	59	89	55	92	38
[Range]	[78–96]	[44–64]	[80–98]	[48–69]	[81–100]	[55–63]	[82–100]	[41–68]	[87–100]	[24–51]

[a]N.A. denotes not applicable, since during winter biogenic volatile organic compound oxidation emissions from vegetation were assumed to be negligible.
[b]Winter periods include the inversion samples.

Protection Agency (EPA) attainment date and was therefore recently reclassified from a moderate to a serious nonattainment area, effective June 2017, becoming one of the first serious nonattainment areas for $PM_{2.5}$ in the U.S.

While much attention has been focused on $PM_{2.5}$ source attribution through analysis of chemical components [*Long et al.*, 2005; *Kelly et al.*, 2013; *Kuprov et al.*, 2014], the sources of the carbonaceous aerosol have remained poorly characterized. However, the carbonaceous aerosol, and in particular the BC fraction, has recently gained increased recognition as a major driver of adverse air pollution health impacts. Several reports have called for consideration of an air pollution health standard for BC, separate from that for $PM_{2.5}$, suggesting that such a standard may prove useful in evaluating local efforts to reduce the population's exposure to combustion PM [*Janssen et al.*, 2012; *Grahame et al.*, 2014]. Here we show that fossil emissions accounted for the majority of BC throughout the year and for more than half of OC during winter, when the air quality was exacerbated by inversions. Under the current state implementation plan for Salt Lake County adopted on 3 December 2014, mobile emissions are the most significant source leading to violation of the $PM_{2.5}$ NAAQS [*Utah State Implementation Plan*, 2014]. This places much of the responsibility for reducing Utah's $PM_{2.5}$ emissions with the federal government, which establishes the emission standards for new motor vehicles. If mobile sources, such as diesel vehicles, are the major contributor to BC and OC emissions, then policy steps to reduce traffic, increase the use of public transportation, and facilitate use of electric vehicles may provide some leverage in mitigating the carbonaceous aerosol concentrations.

Utah has implemented voluntary traffic-reducing actions (for example, reducing vehicle use by consolidating trips) when the Utah DAQ air quality forecasting predicts an inversion period [*Smith and Johnson*, 2013]. However, a study examining the effectiveness of air quality alert systems in Salt Lake County showed that yellow and red air quality alert days were associated with increased traffic in the winter, possibly because more people drive to avoid exposure to the poor air quality and colder temperatures [*Tribby et al.*, 2013]. As a result, improvements in vehicle emissions may be offset by increases in traffic. States have the power to regulate vehicles after they are sold to consumers, but efforts to control vehicle use are difficult to implement due to the large number of vehicles and public opposition to control measures. A positive example of the successful implementation of such air pollution control measures can be seen in California where the transition to lower emission gasoline, a cleaner diesel fleet, and use of electrical energy has significantly reduced PM loads from vehicle and vessel traffic [*Parrish et al.*, 2016].

Area sources (smaller stationary sources, such as home heating, agricultural burning and harvesting, construction, residential and commercial energy generation, wildfires, and biogenic emissions) in the Salt Lake area are the second most important source of $PM_{2.5}$ emissions, and the Utah DAQ listed solid fuel burning as the most important area source of pollution in 2014 [*Utah State Implementation Plan*, 2014]. Among the new area source regulations, a Winter Control Program was implemented, including mandatory solid fuel burning restrictions. Wood and coal burning stoves or fireplaces must not be used when Utah DAQ air quality forecasts predict an inversion period [*Utah State Implementation Plan*, 2014]. However, enforcement of the rule limiting the use of wood as a source of residential heating is challenging. Our findings indicate that contemporary biomass sources, including biomass burning, contributed to nearly half of winter OC during 2013 and 2014. Given the relatively high precision and accuracy of the aerosol ^{14}C measurements, continued monitoring may allow policymakers to identify whether residential wood burning was responding as expected to existing and future regulations.

The recent reclassification of Salt Lake County from a moderate to a serious nonattainment area for $PM_{2.5}$ means that the DAQ must submit a new serious area state implementation plan delineating new control strategies. The development of an improved emissions inventory will be a critical factor in determining the stringency of the requirements described in the plan. In this context, our data set of 31 ^{14}C BC and OC aerosol measurements provides an important baseline for the fossil and contemporary biomass components of carbonaceous aerosol. A future step toward achieving $PM_{2.5}$ emissions consistent with NAAQS would be to use this data set to develop an improved emission inventory.

4.2. Comparison With Other Metropolitan Areas

In the U.S., previous studies have only analyzed the ^{14}C of TC in PM [*Bench*, 2004; *Lewis et al.*, 2004; *Lewis and Stiles*, 2006; *Bench et al.*, 2007; *Schichtel et al.*, 2008]. The results from these studies indicate that contemporary

biomass carbon was predominant in $PM_{2.5}$, accounting for 56–80% in Nashville, Tennessee [*Lewis et al.*, 2004], and 52–89% in Tampa, Florida [*Lewis and Stiles*, 2006]. These studies further recommend that to obtain a better understanding of the sources driving the variations in the carbonaceous aerosol, new ^{14}C measurements of BC and OC fractions are needed.

One major challenge with respect to the successful application of ^{14}C for source apportionment of BC and OC is the complete physical separation of the two fractions without cross-fraction interference and with minimal contamination from background carbon. Aside from the time-consuming nature of these measurements, another limiting factor is the collection of sufficient aerosol mass to prepare targets suitable for the accelerator mass spectrometer [*Szidat et al.*, 2013; *Heal*, 2014].

Here we present, as far as we are aware, the first study in the U.S. which separately evaluates the ^{14}C content of BC and OC aerosols. Our observations indicate that fossil sources are larger than contemporary biomass sources for both BC and OC in Salt Lake City, although the relative contributions are different for each carbon fraction. Similar results have been observed in other metropolitan areas. In Milan, Italy, fossil emissions account for 84% of BC and 37% of OC in the wintertime aerosol [*Bernardoni et al.*, 2013]. In Zurich, Switzerland, on average 75% of BC and 32% of OC during winter has a fossil origin [*Szidat et al.*, 2006]. Significant contributions from fossil sources to BC have also been reported for London (76 to 93% for BC, in contrast to 28 to 46% fossil TC) [*Crilley et al.*, 2015] and for Chinese megacities (75 ± 8% fossil BC, compared to 45 ± 10% fossil OC) [*Zhang et al.*, 2015]. Non-fossil source contributions are typically dominant, however, in areas with greater combustion of biofuels (e.g., India [*Kirillova et al.*, 2014]) or in areas surrounded by evergreen vegetation and subject to wildfire smoke (e.g., Mexico City [*Hodzic et al.*, 2010]).

The higher fossil contribution to BC and OC we observe in Salt Lake metropolitan area is consistent with previous PM ^{14}C observations, which in the U.S. generally have had a higher fossil contribution (50% fossil) than periurban (3–30%) or rural areas (0–18%) [*Bench et al.*, 2007; *Schichtel et al.*, 2008].

4.3. Other Benefits From Reducing BC Emissions in Salt Lake City

4.3.1. Climate

Black carbon is of great scientific and regulatory interest because it is also recognized as the second most important climate warming anthropogenic emission in the present-day atmosphere (after CO_2) [*Ramanathan and Carmichael*, 2008; *Bond et al.*, 2013]. Black carbon is often transported over long distances, mixing with other aerosols along the way, and while sources are often localized, BC can still influence air quality and climate on local, regional, and global scales [*Ramachandran and Kedia*, 2010]. Given that BC has a significant contribution to global radiative forcing, and a much shorter lifetime (on the order of days to weeks) compared with CO_2, the prospect of achieving rapid climate and health benefits by specifically reducing BC emission has attracted much attention [*Grieshop et al.*, 2009; *Arctic Council Task Force on SLCF*, 2011; *U.S. Environmental Protection Agency*, 2012].

Warming from BC aerosols is amplified by deposition on snow and ice surfaces, which reduces surface albedo, increases surface solar heating, and accelerates snow and ice melt [*Wiscombe and Warren*, 1980; *Flanner et al.*, 2007, 2009; *Hadley and Kirchstetter*, 2012; *Lee et al.*, 2013; *Jiao et al.*, 2014]. Winter inversions in the Salt Lake Valley may increase aerosol deposition in nearby snow-covered areas. Anthropogenic air pollution in the Wasatch Range during winter, for example, has been shown to represent a significant source of ion deposition to snow-covered ecosystems [*Hall et al.*, 2014], and carbonaceous matter, in particular, is associated with a significant reduction in snow albedo in the Wasatch Range of northern Utah [*Reynolds et al.*, 2013]. Here we have shown that wintertime BC in Salt Lake Valley originates primarily from fossil fuel combustion. An important next step in this context is to use regional atmospheric models to quantify how BC mitigation in Salt Lake may increase albedo and reduce the melt rate of snow in region, thus slowing rates of regional climate warming.

4.3.2. Health

Apart from complying with the WHO and the NAAQS for $PM_{2.5}$, policy measures focused on achieving the greatest health benefits may require specifically targeting BC reductions. Recent studies have directly related BC to the adverse health effects of air quality and suggested that it is a better indicator of harmful health impacts from combustion-related sources (especially traffic) than PM mass [*Janssen et al.*, 2011, 2012;

Grahame et al., 2014]. This is particularly relevant for the Salt Lake area where short-term exposure to air pollution has been linked to increased rate of emergency department visits for asthma [*Beard et al.*, 2012], a larger number of acute coronary events in people with preexisting conditions [*Pope et al.*, 2015], and increased suicide risk [*Bakian et al.*, 2015].

4.3.3. Visibility

Reducing BC emissions can further contribute to improved visibility and reduced haze. The scenic beauty of Salt Lake City is one of its defining features, with tourism contributing significantly to the local economy [*Leaver*, 2014]. Visibility in urban settings is protected by the secondary PM NAAQS; however, since BC has higher extinction efficiency than most aerosols, reducing BC would have additional visibility benefits over other aerosols in Salt Lake City.

5. Conclusions

Here we used an optimized method for BC-OC separation [*Zhang et al.*, 2012] and ^{14}C analysis of micro-scale aerosol samples [*Mouteva et al.*, 2015a] to estimate fossil and contemporary source contributions to BC and OC in a U.S. city. Our measurements indicate that for Salt Lake City, BC concentrations throughout the year were dominated by fossil fuel emissions, whereas OC had a significant contemporary biomass component that was larger in summer than in winter. Future measurements of ^{14}C may provide information about the efficacy of air quality control measures, which is one of the major goals of the Utah DAQ air-monitoring network. Our results suggest that the use of ^{14}C for monitoring BC and OC aerosols may improve our understanding of source composition in other nonattainment areas, and long-term time series may provide information about the success of different climate and air pollution mitigation policies.

Appendix A

Table A1 provides a detailed summary of all data collected and analyzed in this study.

Table A1. Summary of Aerosol Sample Concentration and Isotopic Composition[a]

Date (Start)	Time (Start) MST	Duration (h)	TC ($\mu gC/m^3$)	BC ($\mu gC/m^3$)	OC ($\mu gC/m^3$)	f_M (TC)	f_M (BC)[b]	f_M (OC)
				Hawthorne				
10/27/2012	12:01 A.M.	47.97	3.9	0.24	3.7	0.573 ± 0.001	0.262 ± 0.039	0.593 ± 0.003
10/29/2012[c]	11:00 A.M.	48.00	6.2	0.77	5.4	0.349 ± 0.038	0.064 (or less)	0.389 ± 0.008[d]
1/8/2013 (inv)	12:00 A.M.	96.00	5.3	0.86	4.4	0.360 ± 0.001	0.118 ± 0.023	0.407 ± 0.005
2/13/2013	2:30 P.M.	48.00	4.8	0.75	4.0	0.367 ± 0.001	0.115 ± 0.052	0.415 ± 0.010
2/16/2013	12:01 A.M.	48.00	3.2	0.34	2.9	0.449 ± 0.001	0.189 ± 0.058	0.480 ± 0.007
2/19/2013	10:00 A.M.	48.00	3.6	0.32	3.3	0.396 ± 0.001	-	-
2/21/2013	10:30 A.M.	24.00	1.8	0.10	1.7	0.649 ± 0.002	-	-
3/13/2013[c]	1:00 P.M.	48.00	2.8	0.16	2.6	0.261 ± 0.022	0.095 (or less)	0.271 ± 0.004[d]
3/26/2013	1:00 P.M.	48.00	2.7	0.14	2.6	0.394 ± 0.001	-	-
4/17/2013	12:05 P.M.	48.00	2.4	0.19	2.2	0.464 ± 0.001	0.119 (or less)	0.493 ± 0.014[d]
4/20/2013	12:01 A.M.	48.00	2.0	0.09	1.9	0.511 ± 0.001	-	-
4/22/2013	10:00 A.M.	48.00	2.1	0.12	2.0	0.483 ± 0.001	0.172 ± 0.046	0.501 ± 0.003
4/24/2013	10:30 A.M.	48.00	4.8	0.27	4.5	0.562 ± 0.001	0.172 ± 0.046	0.586 ± 0.003
6/17/2013	1:00 P.M.	53.00	3.0	0.29	2.7	0.656 ± 0.001	0.057 (or less)	0.720 ± 0.028[d]
8/5/2013	2:17 P.M.	48.00	4.2	0.23	3.9	0.487 ± 0.002	0.105 ± 0.051	0.509 ± 0.003
8/7/2013	3:19 P.M.	47.93	3.8	0.15	3.7	0.588 ± 0.001	0.105 ± 0.051	0.607 ± 0.002
8/9/2013	11:59 P.M.	48.00	2.0	0.07	2.0	0.674 ± 0.002	-	-
8/12/2013	2:37 P.M.	48.00	3.8	0.19	3.6	0.810 ± 0.002	0.109 (or less)	0.847 ± 0.015[d]
10/16/2013	2:45 P.M.	48.25	13.5	0.38	13.1	0.316 ± 0.001	0.116 ± 0.050	0.322 ± 0.002
10/23/2013	2:15 P.M.	49.33	23.7	0.66	23.0	0.448 ± 0.001	0.053 (or less)	0.459 ± 0.001[d]
10/28/2013	3:40 P.M.	50.67	5.2	0.17	5.1	0.539 ± 0.001	0.176 ± 0.054	0.551 ± 0.002
1/1/2014[c] (inv)	-	48.00	5.6	0.60	5.0	0.441 ± 0.024	0.142 ± 0.031	0.477 ± 0.004
1/18/2014 (inv)	-	48.00	8.6	0.37	8.2	0.418 ± 0.001	0.101 (or less)	0.432 ± 0.003[d]
1/21/2014 (inv)	-	48.00	7.9	0.54	7.4	0.364 ± 0.001	0.077 (or less)	0.385 ± 0.003[d]
1/24/2014	-	48.00	17.4	0.36	17.1	0.248 ± 0.001	0.030 (or less)	0.252 ± 0.002[d]
2/1/2014	-	24.00	21.0	0.17	20.8	0.303 ± 0.001	0.157 ± 0.040	0.304 ± 0.001
Average			6.4	0.31	6.0	0.466	0.121	0.476

Table A1. (continued)

Date (Start)	Time (Start) MST	Duration (h)	TC ($\mu gC/m^3$)	BC ($\mu gC/m^3$)	OC ($\mu gC/m^3$)	f_M (TC)	f_M (BC)[b]	f_M (OC)
				Rose Park				
10/31/2012	1:30 P.M.	47.50	3.1	0.23	2.9	0.531 ± 0.002	0.127 ± 0.035	0.563 ± 0.003
2/16/2013	12:01 A.M.	48.00	2.2	0.21	2.0	0.572 ± 0.002	0.276 ± 0.026	0.602 ± 0.003
2/19/2013	11:00 A.M.	48.00	2.1	0.18	1.9	0.395 ± 0.002	0.180 ± 0.059	0.416 ± 0.006
2/21/2013	11:15 A.M.	24.00	2.2	0.18	2.0	0.597 ± 0.002	0.180 ± 0.059	0.635 ± 0.006
4/20/2013	12:01 A.M.	48.00	0.9	-	-	0.571 ± 0.002	-	-
4/22/2013	9:20 A.M.	48.00	1.4	-	-	0.505 ± 0.002	-	-
8/7/2013	2:02 P.M.	48.00	3.0	0.02	3.0	0.618 ± 0.002	0.137 ± 0.068	0.620 ± 0.002
8/9/2013[c]	11:59 P.M.	48.00	1.7	0.13	1.6	0.667 ± 0.002	-	-
Average			2.1	0.16	2.2	0.557	0.180	0.567
				University of Utah				
10/24/2012	12:00 P.M.	48.00	1.5	0.09	1.4	0.518 ± 0.001	-	-
10/27/2012	12:01 A.M.	47.97	1.7	0.07	1.7	0.567 ± 0.001	-	-
10/29/2012	11:15 A.M.	48.00	2.4	0.14	2.2	0.382 ± 0.001	0.180 ± 0.066	0.395 ± 0.004
10/31/2012	2:30 P.M.	47.00	2.2	0.11	2.1	0.518 ± 0.001	0.180 ± 0.066	0.535 ± 0.004
2/13/2013	2:00 P.M.	48.00	2.5	0.29	2.3	0.339 ± 0.001	0.081 (or less)	0.372 ± 0.009[d]
4/17/2013	11:20 A.M.	48.00	0.8	-	-	0.442 ± 0.002	-	-
8/5/2013	12:17 P.M.	46.22	2.4	0.15	2.2	0.493 ± 0.001	0.098 (or less)	0.519 ± 0.004[d]
Average			1.9	0.14	2.0	0.466	0.134	0.455

[a]Periods of inversion are denoted with "(inv)" by the start date of the sample. Dates are formatted as month/day/year.
[b]Several BC samples reached the AMS measurement limit, and their f_M is reported as a given value or smaller. The true value can be as low as 0.
[c]Samples which were duplicated in the ^{14}C analysis of TC. Reported values for TC concentrations and f_M(TC) are the average and standard deviation of the duplicates.
[d]The corresponding f_M(OC) of samples reaching f_M(BC) limit were calculated assuming that the reported n(BC) was correct and propagating the error as a square root.

References

Ackerman, A., O. Toon, J. Taylor, D. Johnson, P. Hobbs, and R. Ferek (2000), Effects of aerosols on cloud albedo: Evaluation of Twomey's parameterization of cloud susceptibility using measurements of ship tracks, *J. Atmos. Sci.*, *1995*, 2684–2695.

Andersson, A., J. Deng, K. Du, M. Zheng, C. Yan, M. Sköld, and Ö. Gustafsson (2015), Regionally-varying combustion sources of the January 2013 severe haze events over eastern China, *Environ. Sci. Technol.*, *49*(4), 2038–2043, doi:10.1021/es503855e.

Arctic Council Task Force on SLCF (2011), An assessment of emissions and mitigation options for black carbon for the Arctic council.

Bakian, A. V., R. S. Huber, H. Coon, D. Gray, P. Wilson, W. M. McMahon, and P. F. Renshaw (2015), Acute air pollution exposure and risk of suicide completion, *Am. J. Epidemiol.*, *181*(5), 295–303, doi:10.1093/aje/kwu341.

Barrett, T. E., E. M. Robinson, S. Usenko, and R. J. Sheesley (2015), Source contributions to wintertime elemental and organic carbon in the western Arctic based on radiocarbon and tracer apportionment, *Environ. Sci. Technol.*, *49*, 11,631–11,639, doi:10.1021/acs.est.5b03081.

Bauer, S. E., S. Menon, D. Koch, T. C. Bond, and K. Tsigaridis (2010), A global modeling study on carbonaceous aerosol microphysical characteristics and radiative effects, *Atmos. Chem. Phys.*, *10*(15), 7439–7456, doi:10.5194/acp-10-7439-2010.

Beard, J. D., C. Beck, R. Graham, S. C. Packham, M. Traphagan, R. T. Giles, and J. G. Morgan (2012), Winter temperature inversions and emergency department visits for asthma in Salt Lake County, Utah, 2003–2008, *Environ. Health Perspect.*, *120*(10), 1385–1390, doi:10.1289/ehp.1104349.

Bench, G. (2004), Measurement of contemporary and fossil carbon contents of PM$_{2.5}$ aerosols: Results from Turtleback Dome, Yosemite National Park, *Environ. Sci. Technol.*, *38*(8), 2424–2427.

Bench, G., S. Fallon, B. Schichtel, W. Malm, and C. McDade (2007), Relative contributions of fossil and contemporary carbon sources to PM$_{2.5}$ aerosols at nine Interagency Monitoring for Protection of Visual Environments (IMPROVE) network sites, *J. Geophys. Res.*, *112*, D10205, doi:10.1029/2006JD007708.

Bernardoni, V., et al. (2013), Radiocarbon analysis on organic and elemental carbon in aerosol samples and source apportionment at an urban site in northern Italy, *J. Aerosol Sci.*, *56*, 88–99, doi:10.1016/j.jaerosci.2012.06.001.

Bond, T. C., D. G. Streets, K. F. Yarber, S. M. Nelson, J. H. Woo, and Z. Klimont (2004), A technology-based global inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.*, *109*, D14203, doi:10.1029/2003JD003697.

Bond, T. C., E. Bhardwaj, R. Dong, R. Jogani, S. Jung, C. Roden, D. G. Streets, and N. M. Trautmann (2007), Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, *Global Biogeochem. Cycles*, *21*, GB2018, doi:10.1029/2006GB002840.

Bond, T. C., et al. (2013), Bounding the role of black carbon in the climate system: A scientific assessment, *J. Geophys. Res. Atmos.*, *118*, 5380–5552, doi:10.1002/jgrd.50171.

Brook, R. D., et al. (2010), Particulate matter air pollution and cardiovascular disease: An update to the scientific statement from the American Heart Association, *Circulation*, *121*(21), 2331–2378, doi:10.1161/CIR.0b013e3181dbece1.

Budhavant, K., A. Andersson, C. Bosch, M. Kruså, E. N. Kirillova, R. J. Sheesley, P. D. Safai, P. S. P. Rao, and Ö. Gustafsson (2015), Radiocarbon-based source apportionment of elemental carbon aerosols at two south Asian receptor observatories over a full annual cycle, *Environ. Res. Lett.*, *10*(6), 064004, doi:10.1088/1748-9326/10/6/064004.

Cao, C., X. Lee, S. Liu, N. Schultz, W. Xiao, M. Zhang, and L. Zhao (2016), Urban heat islands in China enhanced by haze pollution, *Nat. Commun.*, *7*, 12509, doi:10.1038/ncomms12509.

Chen, B., et al. (2013), Source forensics of black carbon aerosols from China, *Environ. Sci. Technol.*, *47*, 9102–9108, doi:10.1021/es401599r.

Chen, H., et al. (2017), Living near major roads and the incidence of dementia, Parkinson's disease, and multiple sclerosis: A population-based cohort study, *Lancet*, *6736*(16), 1–11, doi:10.1016/S0140-6736(16)32399-6.

Cozic, J., B. Verheggen, S. Mertes, P. Connolly, K. Bower, A. Petzold, U. Baltensperger, and E. Weingartner (2007), Scavenging of black carbon in mixed phase clouds at the high alpine site Jungfraujoch, *Atmos. Chem. Phys.*, *7*(7), 1797–1807, doi:10.5194/acp-7-1797-2007.

Crilley, L. R., et al. (2015), Sources and contributions of wood smoke during winter in London: Assessing local and regional influences, *Atmos. Chem. Phys.*, *15*(6), 3149–3171, doi:10.5194/acp-15-3149-2015.

Dockery, D. W., and C. A. Pope (1994), Acute respiratory effects of particulate air pollution, *Annu. Rev. Public Health*, *15*, 107–132, doi:10.1146/annurev.pu.15.050194.000543.

Dominici, F., R. D. Peng, M. L. Bell, L. Pham, A. McDermott, S. L. Zeger, and J. M. Samet (2006), Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases, *JAMA J. Am. Med. Assoc. Am. Med. Assoc.*, *295*(10), 1127–1134, doi:10.1001/jama.295.10.1127.

Fann, N., A. D. Lamson, S. C. Anenberg, K. Wesson, D. Risley, and B. J. Hubbell (2012), Estimating the national public health burden associated with exposure to ambient $PM_{2.5}$ and ozone, *Risk Anal.*, *32*(1), 81–95, doi:10.1111/j.1539-6924.2011.01630.x.

Flanner, M. G., C. S. Zender, J. T. Randerson, and P. J. Rasch (2007), Present-day climate forcing and response from black carbon in snow, *J. Geophys. Res.*, *112*, D11202, doi:10.1029/2006JD008003.

Flanner, M. G., C. S. Zender, P. G. Hess, N. M. Mahowald, T. H. Painter, V. Ramanathan, and P. J. Rasch (2009), Springtime warming and reduced snow cover from carbonaceous particles, *Atmos. Chem. Phys.*, *9*(7), 2481–2497, doi:10.5194/acp-9-2481-2009.

Gelencser, A. (2004), *Carbonaceous Aerosol*, edited by K. Mysak and L. A. Hamilton, Springer, Netherlands.

Gelencsér, A., B. May, D. Simpson, A. Sánchez-Ochoa, A. Kasper-Giebl, H. Puxbaum, A. Caseiro, C. A. Pio, and M. Legrand (2007), Source apportionment of $PM_{2.5}$ organic aerosol over Europe: Primary/secondary, natural/anthropogenic, and fossil/biogenic origin, *J. Geophys. Res.*, *112*, D23S04, doi:10.1029/2006JD008094.

Gillies, R. R., S.-Y. Wang, and M. R. Booth (2010), Atmospheric scale interaction on wintertime intermountain west low-level inversions, *Weather Forecasting*, *25*(4), 1196–1210, doi:10.1175/2010WAF2222380.1.

Goldstein, A. H., and I. E. Galbally (2007), Known and unexplored organic constituents in the Earth's atmosphere, *Environ. Sci. Technol.*, *41*(5), 1514–1521, doi:10.1021/es072476p.

Götschi, T., J. Heinrich, J. Sunyer, and N. Künzli (2008), Long-term effects of ambient air pollution on lung function: A review, *Epidemiology*, *19*(5), 690–701, doi:10.1097/EDE.0b013e318181650f.

Grahame, T. J., R. Klemm, and R. B. Schlesinger (2014), Public health and components of particulate matter: The changing assessment of black carbon, *J. Air Waste Manage. Assoc.*, *64*(6), 620–660, doi:10.1080/10962247.2014.912692.

Grieshop, A. P., C. C. O. Reynolds, M. Kandlikar, and H. Dowlatabadi (2009), A black-carbon mitigation wedge, *Nat. Geosci.*, *2*(8), 533–534, doi:10.1038/ngeo595.

Griffin, R. J., R. Cocker, and C. Flagan (1999), Organic aerosol formation from the oxidation of biogenic hydrocarbons, *J. Geophys. Res.*, *104*(1998), 3555–3567, doi:10.1029/1998JD100049.

Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, and C. Geron (2006), Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, *6*(11), 3181–3210.

Gustafsson, O., M. Kruså, Z. Zencak, R. J. Sheesley, L. Granat, E. Engström, P. S. Praveen, P. S. P. Rao, C. Leck, and H. Rodhe (2009), Brown clouds over South Asia: Biomass or fossil fuel combustion?, *Science*, *323*(5913), 495–498, doi:10.1126/science.1164857.

Hadley, O. L., and T. W. Kirchstetter (2012), Black-carbon reduction of snow albedo, *Nat. Clim. Change*, *2*(6), 437–440, doi:10.1038/nclimate1433.

Hall, S. J., G. Maurer, S. W. Hoch, R. Taylor, and D. R. Bowling (2014), Impacts of anthropogenic emissions and cold air pools on urban to montane gradients of snowpack ion concentrations in the Wasatch Mountains, Utah, *Atmos. Environ.*, *98*, 231–241, doi:10.1016/j.atmosenv.2014.08.076.

Hansen, M. C., et al. (2013), High-resolution global maps of 21st-century Forest cover change, *Science*, *342*(6160), 850–853, doi:10.1126/science.1244693.

Haywood, J., and O. Boucher (2000), Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review, *Rev. Geophys.*, *38*(4), 513–543, doi:10.1029/1999RG000078.

Heal, M. R. (2014), The application of carbon-14 analyses to the source apportionment of atmospheric carbonaceous particulate matter: A review, *Anal. Bioanal. Chem.*, *406*(1), 81–98, doi:10.1007/s00216-013-7404-1.

Hodzic, A., J. L. Jimenez, A. S. H. Prévôt, S. Szidat, J. D. Fast, and S. Madronich (2010), Can 3-D models explain the observed fractions of fossil and non-fossil carbon in and near Mexico City?, *Atmos. Chem. Phys.*, *10*(22), 10,997–11,016, doi:10.5194/acp-10-10997-2010.

Janssen, N. A., M. E. Gerlofs-Nijland, T. Lanki, R. O. Salonen, F. Cassee, G. Hoek, P. Fischer, B. Brunekreef, and M. Krzyzanowski (2012), *Health Effects of Black Carbon*, WHO Regional Office for Europe, Copenhagen, Denmark.

Janssen, N. A. H., et al. (2011), Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM_{10} and $PM_{2.5}$, *Environ. Health Perspect.*, *12*, 1691–1699.

Jiao, C., et al. (2014), An AeroCom assessment of black carbon in Arctic snow and sea ice, *Atmos. Chem. Phys.*, *14*(5), 2399–2417, doi:10.5194/acp-14-2399-2014.

Johnston, F. H., S. B. Henderson, Y. Chen, J. T. Randerson, M. Marlier, R. S. Defries, P. Kinney, D. M. J. S. Bowman, and M. Brauer (2012), Estimated global mortality attributable to smoke from landscape fires, *Environ. Health Perspect.*, *120*(5), 695–701, doi:10.1289/ehp.1104422.

Junker, C., and C. Liousse (2006), A global emission inventory of carbonaceous aerosol from historic records of fossil fuel and biofuel consumption for the period 1860–1997, *Atmos. Chem. Phys. Discuss.*, *6*(3), 4897–4927, doi:10.5194/acpd-6-4897-2006.

Kanakidou, M., et al. (2005), Organic aerosol and global climate modelling: A review, *Atmos. Chem. Phys.*, *5*(4), 1053–1123, doi:10.5194/acp-5-1053-2005.

Kelly, R., M. L. Chipman, P. E. Higuera, I. Stefanova, L. B. Brubaker, and F. S. Hu (2013), Recent burning of boreal forests exceeds fire regime limits of the past 10,000 years, *Proc. Natl. Acad. Sci. U.S.A.*, *110*(32), 13,055–13,060, doi:10.1073/pnas.1305069110.

Kesselmeier, J., and M. Staudt (1999), Biogenic volatile organic compounds (VOC): An overview on emission, physiology and ecology, *J. Atmos. Chem.*, *33*(1), 23–88, doi:10.1023/A:1006127516791.

Kirillova, E. N., A. Andersson, S. Tiwari, A. Kumar Srivastava, D. Singh Bisht, and Ö. Gustafsson (2014), Water-soluble organic carbon aerosols during a full New Delhi winter: Isotope-base source apportionment and optical properties, *J. Geophys. Res. Atmos.*, *119*, 3476–3485, doi:10.1002/2013JD021272.Received.

Kloog, I., A. Zanobetti, F. Nordio, B. A. Coull, A. A. Baccarelli, and J. Schwartz (2015), Effects of airborne fine particles ($PM_{2.5}$) on deep vein thrombosis admissions in north eastern United States, *J. Thromb. Haemost.*, 768–774, doi:10.1111/jth.12873.

Knaapen, A. M., P. J. A. Borm, C. Albrecht, and R. P. F. Schins (2004), Inhaled particles and lung cancer. Part A: Mechanisms, *Int. J. Cancer*, *109*(6), 799–809, doi:10.1002/ijc.11708.

Koch, D., and A. D. Del Genio (2010), Black carbon semi-direct effects on cloud cover: Review and synthesis, *Atmos. Chem. Phys.*, *10*(16), 7685–7696, doi:10.5194/acp-10-7685-2010.

Kuprov, R., D. J. Eatough, T. Cruickshank, N. Olson, P. M. Cropper, and J. C. Hansen (2014), Composition and secondary formation of fine particulate matter in the Salt Lake Valley: Winter 2009, *J. Air Waste Manage. Assoc.*, *64*(8), 957–969, doi:10.1080/10962247.2014.903878.

Leaver, J. (2014), The state of Utah's tourism, travel and recreation industry, *Utah Econ. Bus. Rev.*, *73*(4), 1–16.

Lee, Y. H., et al. (2013), Evaluation of preindustrial to present-day black carbon and its albedo forcing from Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, *13*(5), 2607–2634, doi:10.5194/acp-13-2607-2013.

Lelieveld, J., J. S. Evans, M. Fnais, D. Giannadaki, and A. Pozzer (2015), The contribution of outdoor air pollution sources to premature mortality on a global scale, *Nature*, *525*(7569), 367–371, doi:10.1038/nature15371.

Lerdau, M., A. Guenther, and R. Monson (2013), Plant production emission of volatile organic compounds plant-produced hydrocarbons influence not only the plant itself but the atmosphere as well, *Bioscience*, *47*(6), 373–383.

Levin, I., T. Naegler, B. Kromer, M. Diehl, R. J. Francey, A. J. Gomez-Pelaez, L. P. Steele, D. Wagenbach, R. Weller, and D. E. Worthy (2010), Observations and modelling of the global distribution and long-term trend of atmospheric $^{14}CO_2$, *Tellus B*, *62*(1), 26–46, doi:10.1111/j.1600-0889.2009.00446.x.

Lewis, C. W., and D. C. Stiles (2006), Radiocarbon content of $PM_{2.5}$ ambient aerosol in Tampa, FL, *Aerosol Sci. Technol.*, *40*(3), 189–196, doi:10.1080/02786820500521007.

Lewis, C. W., G. A. Klouda, and W. D. Ellenson (2004), Radiocarbon measurement of the biogenic contribution to summertime $PM_{2.5}$ ambient aerosol in Nashville, TN, *Atmos. Environ.*, *38*(35), 6053–6061, doi:10.1016/j.atmosenv.2004.06.011.

Long, R. W., et al. (2005), One- and three–hour $PM_{2.5}$ characterization, speciation, and source apportionment using continuous and integrated samplers, *Aerosol Sci. Technol.*, *39*(3), 238–248, doi:10.1080/027868290925633.

Miller, K., D. Siscovick, L. Sheppard, K. Shepherd, G. A. SullivanJeffrey, and J. D. Kaufman (2007), Long-term exposure to air pollution and incidence of cardiovascular events in women, *N. Engl. J. Med.*, *356*(5), 447–458, doi:10.1056/NEJMoa1005372.

Mouteva, G. O., S. M. Fahrni, G. M. Santos, J. T. Randerson, Y. L. Zhang, S. Szidat, and C. I. Czimczik (2015a), Accuracy and precision of ^{14}C-based source apportionment of organic and elemental carbon in aerosols using the Swiss_4S protocol, *Atmos. Meas. Tech.*, *8*(4), 3729–3743, doi:10.5194/amtd-8-3933-2015.

Mouteva, G. O., et al. (2015b), Black carbon aerosol dynamics and isotopic composition in Alaska linked with boreal fire emissions and depth of burn in organic soils, *Global Biogeochem. Cycles*, *29*, 1977–2000, doi:10.1002/2015GB005247.

Myhre, G., et al. (2013), 2013: Anthropogenic and natural radiative forcing, in *Clim. Chang. 2013 Phys. Sci. Basis. Contrib. Work. Gr. I to Fifth Assess. Rep. Intergov. Panel Clim. Chang.*, chap. 8, pp. 659–740, Cambridge Univ. Press, Cambridge, U. K., and New York, doi:10.1017/CBO9781107415324.018.

Novakov, T. (2003), Large historical changes of fossil-fuel black carbon aerosols, *Geophys. Res. Lett.*, *30*(6), 1324, doi:10.1029/2002GL016345.

Ostro, B., R. Broadwin, S. Green, W. Y. Feng, and M. Lipsett (2006), Fine particulate air pollution and mortality in nine California counties: Results from CALFINE, *Environ. Health Perspect.*, *114*(1), 29–33, doi:10.1289/ehp.8335.

Oudin, A., B. Forsberg, A. N. Adolfsson, N. Lind, L. Modig, M. Nordin, S. Nordin, R. Adolfsson, and L. G. Nilsson (2016), Traffic-related air pollution and dementia incidence in northern Sweden: A longitudinal study, *Environ. Health Perspect.*, *124*(3), 306–312, doi:10.1289/ehp.1408322.

Parrish, D. D., J. Xu, B. Croes, and M. Shao (2016), Air quality improvement in Los Angeles—Perspectives for developing cities, *Front. Environ. Sci. Eng.*, *10*(5), doi:10.1007/s11783-016-0859-5.

Pataki, D. E., D. R. Bowling, and J. R. Ehleringer (2003), Seasonal cycle of carbon dioxide and its isotopic composition in an urban atmosphere: Anthropogenic and biogenic effects, *J. Geophys. Res.*, *108*(D23), 4735, doi:10.1029/2003JD003865.

Penner, J. (1995), Carbonaceous aerosols influencing atmospheric radiation: Black and organic carbon, in *Aerosol Forcing of Climate: Report of the Dahlem Workshop on Aerosol Forcing of Climate, Berlin 1994, April 24–29*, edited by R. J. Charlston and J. Heintzenberg, pp. 91–108, John Wiley, Chichester, New York.

Penner, J. E., and T. Novakov (1996), Carbonaceous particles in the atmosphere: A historical perspective to the Fifth International Conference on Carbonaceous Particles in the Atmosphere, *J. Geophys. Res.*, *101*(D14), 19,373–19,378, doi:10.1029/96JD01175.

Penner, J. E., M. J. Prather, I. S. A. Isaksen, J. S. Fuglestvedt, Z. Klimont, and D. S. Stevenson (2010), Short-lived uncertainty?, *Nat. Geosci.*, *3*(9), 587–588, doi:10.1038/ngeo932.

Pope, C. A., R. T. Burnett, M. C. Turner, A. Cohen, D. Krewski, M. Jerrett, S. M. Gapstur, and M. J. Thun (2011), Lung cancer and cardiovascular disease mortality associated with ambient air pollution and cigarette smoke: Shape of the exposure-response relationships, *Environ. Health Perspect.*, *119*(11), 1616–1621, doi:10.1289/ehp.1103639.

Pope, C. A., J. B. Muhlestein, J. L. Anderson, J. B. Cannon, N. M. Hales, K. G. Meredith, V. Le, and B. D. Horne (2015), Short-term exposure to fine particulate matter air pollution is preferentially associated with the risk of ST-segment elevation acute coronary events, *J. Am. Heart Assoc.*, *4*(12), e002506, doi:10.1161/JAHA.115.002506.

Ramachandran, S., and S. Kedia (2010), Black carbon aerosols over an urban region: Radiative forcing and climate impact, *J. Geophys. Res.*, *115*, D10202, doi:10.1029/2009JD013560.

Ramanathan, V., and G. Carmichael (2008), Global and regional climate changes due to black carbon, *Nat. Geosci.*, *I*, 221–227.

Reddy, C. M., A. Pearson, L. Xu, A. P. McNichol, B. A. Benner, S. A. Wise, G. A. Klouda, L. A. Currie, and T. I. Eglinton (2002), Radiocarbon as a tool to apportion the sources of polycyclic aromatic hydrocarbons and black carbon in environmental samples, *Environ. Sci. Technol.*, *36*(8), 1774–1782.

Reynolds, R. L., et al. (2013), Composition of dust deposited to snow cover in the Wasatch Range (Utah, USA): Controls on radiative properties of snow cover and comparison to some dust-source sediments, *Aeolian Res.*, *15*, 73–90, doi:10.1016/j.aeolia.2013.08.001.

Rice, M. B., P. L. Ljungman, E. H. Wilker, K. S. Dorans, D. R. Gold, J. Schwartz, P. Koutrakis, G. R. Washko, G. T. O'Connor, and M. A. Mittleman (2015), Long-term exposure to traffic emissions and fine particulate matter and lung function decline in the Framingham Heart Study, *Am. J. Respir. Crit. Care Med.*, *191*(6), 656–664, doi:10.1164/rccm.201410-1875OC.

Rosenfeld, D. (2000), Suppression of rain and snow by urban and industrial air pollution, *Science*, *287*(5459), 1793–1796, doi:10.1126/science.287.5459.1793.

Santos, G. M., J. R. Southon, S. Griffin, S. R. Beaupre, and E. R. M. Druffel (2007), Ultra small-mass AMS ^{14}C sample preparation and analyses at KCCAMS/UCI Facility, *Nucl. Instrum. Methods Phys. Res., Sect. B*, *259*(1), 293–302, doi:10.1016/j.nimb.2007.01.172.

Schichtel, B. A., W. C. Malm, G. Bench, S. Fallon, C. E. McDade, J. C. Chow, and J. G. Watson (2008), Fossil and contemporary fine particulate carbon fractions at 12 rural and urban sites in the United States, *J. Geophys. Res.*, *113*, D02311, doi:10.1029/2007JD008605.

Schwartz, J., D. W. Dockery, and L. M. Neas (1996), Is daily mortality associated specifically with fine particles?, *J. Air Waste Manage. Assoc.*, *46*(10), 927–939, doi:10.1080/10473289.1996.10467528.

Sheppard, L., D. Levy, G. Norris, T. Larson, and J. Koenig (1999), Effects of ambient air pollution on nonelderly asthma hospital admissions in Seattle, Washington, *Epidemiology*, *10*(1), 23–30, doi:10.1097/00001648-200509000-00189.

Silcox, G. D., K. E. Kelly, E. T. Crosman, C. D. Whiteman, and B. L. Allen (2012), Wintertime $PM_{2.5}$ concentrations during persistent, multi-day cold-air pools in a mountain valley, *Atmos. Environ.*, *46*, 17–24, doi:10.1016/j.atmosenv.2011.10.041.

Slater, J. F., L. A. Currie, J. E. Dibb, and B. A. Benner (2002), Distinguishing the relative contribution of fossil fuel and biomass combustion aerosols deposited at Summit, Greenland through isotopic and molecular characterization of insoluble carbon, *Atmos. Environ.*, *36*, 4463–4477, doi:10.1016/S1352-2310(02)00402-8.

Smith, A., and B. Johnson (2013), News release: DAQ launches new alert system for wood burning restrictions,

Stone, E. A., G. C. Lough, J. J. Schauer, P. S. Praveen, C. E. Corrigan, and V. Ramanathan (2007), Understanding the origin of black carbon in the atmospheric brown cloud over the Indian Ocean, *J. Geophys. Res.*, *112*, D22S23, doi:10.1029/2006JD008118.

Stuiver, M., and H. Polach (1977), Discussion: Reporting of ^{14}C data, *Radiocarbon*, *19*(3), 355–363.

Suess, H. E. (1955), Radiocarbon concentration in modern wood, *Science*, *122*(3166), 415–417, doi:10.1126/science.122.3166.415-a.

Szidat, S., T. M. Jenk, H.-A. Synal, M. Kalberer, L. Wacker, I. Hajdas, A. Kasper-Giebl, and U. Baltensperger (2006), Contributions of fossil fuel, biomass-burning, and biogenic emissions to carbonaceous aerosols in Zurich as traced by ^{14}C, *J. Geophys. Res.*, *111*, D07206, doi:10.1029/2005JD006590.

Szidat, S., A. S. H. Prévôt, J. Sandradewi, M. R. Alfarra, H. A. Synal, L. Wacker, and U. Baltensperger (2007), Dominant impact of residential wood burning on particulate matter in Alpine valleys during winter, *Geophys. Res. Lett.*, *34*, L05820, doi:10.1029/2006GL028325.

Szidat, S., M. Ruff, N. Perron, L. Wacker, H. A. Synal, M. Hallquist, A. S. Shannigrahi, K. E. Yttri, C. Dye, and D. Simpson (2009), Fossil and non-fossil sources of organic carbon (OC) and elemental carbon (EC) in Göteborg, Sweden, *Atmos. Chem. Phys.*, *9*, 1521–1535.

Szidat, S., et al. (2013), ^{14}C analysis of carbonaceous aerosols: Exercise 2009, *Radiocarbon*, *55*(2–3), 1496–1509.

Tribby, C. P., H. J. Miller, Y. Song, and K. R. Smith (2013), Do air quality alerts reduce traffic? An analysis of traffic data from the Salt Lake City metropolitan area, Utah, USA, *Transp. Policy*, *30*, 173–185, doi:10.1016/j.tranpol.2013.09.012.

U.S. Census Bureau, P. D. (2017), Annual estimates of the resident population: April 1, 2010 to July 1, 2016. [Available at http://factfinder. census.gov/faces/tableservices/jsf/pages/productview.xhtml?src=bkmk.]

U.S. Environmental Protection Agency (2012), Report to congress on black carbon.

Utah Division of Air Quality (2015), Utah division of air quality 2015 annual report, Salt Lake City.

Utah State Implementation Plan (2014), Control measures for area and point sources, fine particulate matter, $PM_{2.5}$ *SIP for the Salt Lake City, UT Nonattainment Area, Salt Lake City*.

Vignati, E., M. Karl, M. Krol, J. Wilson, P. Stier, and F. Cavalli (2010), Sources of uncertainties in modelling black carbon at the global scale, *Atmos. Chem. Phys.*, *10*(6), 2595–2611, doi:10.5194/acpd-9-24317-2009.

Van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, M. Mu, P. S. Kasibhatla, D. C. Morton, R. S. DeFries, Y. Jin, and T. T. van Leeuwen (2010), Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmos. Chem. Phys.*, *10*(23), 11,707–11,735, doi:10.5194/acp-10-11707-2010.

West, J., et al. (2016), What we breathe impacts our health: Improving understanding of the link between air pollution and health, *Environ. Sci. Technol.*, acs.est.5b03827, doi:10.1021/acs.est.5b03827.

Whiteman, C. D., S. W. Hoch, J. D. Horel, and A. Charland (2014), Relationship between particulate air pollution and meteorological variables in Utah's Salt Lake Valley, *Atmos. Environ.*, *94*, 742–753, doi:10.1016/j.atmosenv.2014.06.012.

Winiger, P., A. Andersson, S. Eckhardt, A. Stohl, and Ö. Gustafsson (2016), The sources of atmospheric black carbon at a European gateway to the Arctic, *Nat. Commun.*, *7*, 12,776, doi:10.1038/ncomms12776.

Winiger, P., et al. (2017), Siberian Arctic black carbon sources constrained by model and observation, *Proc. Natl. Acad. Sci. U.S.A.*, *114*(7), E1054–E1061, doi:10.1073/pnas.1613401114.

Wiscombe, J. W., and S. G. Warren (1980), A model for the spectral albedo of snow. I: Pure snow, *J. Atmos. Sci.*, *37*, 2712–2733.

Xu, X., S. E. Trumbore, S. Zheng, J. R. Southon, K. E. McDuffee, M. Luttgen, and J. C. Liu (2007), Modifying a sealed tube zinc reduction method for preparation of AMS graphite targets: Reducing background and attaining high precision, *Nucl. Instrum. Methods Phys. Res., Sect. B*, *259*(1), 320–329, doi:10.1016/j.nimb.2007.01.175.

Young, M. T., D. P. Sandler, L. A. Deroo, S. Vedal, J. D. Kaufman, and S. J. London (2014), Ambient air pollution exposure and incident adult asthma in a nationwide cohort of U.S. women, *Am. J. Respir. Crit. Care Med.*, *190*(8), 914–921, doi:10.1164/rccm.201403-0525OC.

Zhang, Y. L., N. Perron, V. G. Ciobanu, P. Zotter, M. C. Minguillón, L. Wacker, A. S. H. Prévôt, U. Baltensperger, and S. Szidat (2012), On the isolation of OC and EC and the optimal strategy of radiocarbon-based source apportionment of carbonaceous aerosols, *Atmos. Chem. Phys.*, *12*(22), 10,841–10,856, doi:10.5194/acp-12-10841-2012.

Zhang, Y. L., et al. (2015), Fossil vs. non-fossil sources of fine carbonaceous aerosols in four Chinese cities during the extreme winter haze episode of 2013, *Atmos. Chem. Phys.*, *15*(3), 1299–1312, doi:10.5194/acp-15-1299-2015.