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Urban impacts on regional carbonaceous aerosols: Case study in central Texas

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Rural and background sites provide valuable information on the concentration and optical properties of organic, elemental, and water-soluble organic carbon (OC, EC, and WSOC), which are relevant for understanding the climate forcing potential of regional atmospheric aerosols. To quantify climate- and air quality-relevant characteristics of carbonaceous aerosol in the central United States, a regional background site in central Texas was chosen for long-term measurement. Back trajectory (BT) analysis, ambient OC, EC, and WSOC concentrations and absorption parameters are reported for the first 15 months of a long-term campaign (May 2011–August 2012). BT analysis indicates consistent north–south airflow connecting central Texas to the Central Plains. Central Texas aerosols exhibited seasonal trends with increased fine particulate matter (<2.5 μm aerodynamic diameter, $PM_{2.5}$) and OC during the summer ($PM_{2.5} = 10.9\ \mu g\ m^{-3}$ and $OC = 3.0\ \mu g\ m^{-3}$) and elevated EC during the winter ($0.22\ \mu g\ m^{-3}$). When compared to measurements in Dallas and Houston, TX, central Texas OC appears to have mixed urban and rural sources. However, central Texas EC appears to be dominated by transport of urban emissions. WSOC averaged 63% of the annual OC, with little seasonal variability in this ratio. To monitor brown carbon (BrC), absorption was measured for the aqueous WSOC extracts. Light absorption coefficients for EC and BrC were highest during summer (EC MAC = $11\ m^2\ g^{-1}$ and BRC $MAE_{365} = 0.15\ m^2\ g^{-1}$). Results from optical analysis indicate that regional aerosol absorption is mostly due to EC with summertime peaks in BrC attenuation. This study represents the first reported values of WSOC absorption, MAE_{365}, for the central United States.

Implications: Background concentration and absorption measurements are essential in determining regional potential radiative forcing due to atmospheric aerosols. Back trajectory, chemical, and optical analysis of $PM_{2.5}$ was used to determine climatic and air quality implications of urban outflow to a regional receptor site, representative of the central United States. Results indicate that central Texas organic carbon has mixed urban and rural sources, while elemental carbon is controlled by the transport of urban emissions. Analysis of aerosol absorption showed black carbon as the dominant absorber, with less brown carbon absorption than regional studies in California and the southeastern United States.

Introduction

Optical properties of aerosols are important in determining climatic impacts on both regional and global scales (Ram and Sarin, 2009). Elemental carbon (EC, equivalent to black carbon) is thought to dominate light absorption by aerosols and is most efficient at absorbing visible light (Bond et al., 2004), with a climate forcing value of +1.1 W m^{-2}, with 90% uncertainty bounds of +0.17 to +2.1 W m^{-2}, resulting in a net global warming of up to 0.8°C (Chung et al., 2005; Chung and Seinfeld, 2005; Ramanathan and Carmichael, 2008; Chen et al., 2012; Bond et al., 2013). Among atmospheric pollutants it is second only to CO_2 in potential forcing (Bond et al., 2013). It has also been proposed that atmospheric heating as a result of EC may affect large-scale circulation and the hydrological cycle, leading to significant regional climate effects (Cheng et al., 2011). Typically, EC is treated as the only light-absorbing species in climate models (Kirchstetter et al., 2004; Cheng et al., 2011). However, the effects of light-absorbing organic brown carbon, or BrC, are becoming more apparent and have been reported at 28% of total absorption at lower wavelengths for California (Doherty et al., 2010; Bahadur et al., 2012). BrC absorbs light from the ultraviolet (UV) to lower visible wavelengths, with absorption greatly increasing as wavelength decreases. Sources of BrC include primary emissions (e.g., incomplete and smoldering combustion), as well as products of photochemical reactions in the atmosphere (Hecobian et al., 2010; Zhang et al., 2011; Zhang et al., 2013). The presence of BrC in atmospheric particulate matter is often determined through the spectral properties of water-soluble organic carbon (WSOC) in aqueous solutions; however, not all WSOC is BrC (Hoffer, 2006). BrC can also be present in the water-insoluble fraction of organic carbon. For this study, BrC refers only to water-soluble, light-absorbing organic carbon.

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Absorption due to carbonaceous aerosols, both the organic and elemental fractions, is currently one of the greatest sources of uncertainty in the global radiative budget (Anderson et al., 2003; McComiskey et al., 2008; Chen and Bond, 2010). Absorption coefficient for WSOC absorption (b_{abs}) and mass absorption cross-section measurements for the United States have been made in the Southeast and on the West Coast, including Atlanta, GA ($b_{abs} = 0.61 \pm 0.38\ M m^{-1}$), and Pasadena, CA ($b_{abs} = 0.88 \pm 0.71\ M m^{-1}$), but are needed for the central United States, particularly for BrC (Hecobian et al., 2010; Zhang et al., 2011). Aerosol absorption measurements are needed in this region to improve regional accuracy and reduce uncertainty in global climate models (Bond and Bergstrom, 2006; Cheng et al., 2011; Thompson et al., 2012).

To characterize carbonaceous aerosols in the central United States, a rural background site in central Texas was chosen. Central Texas is frequently impacted by atmospheric pollution from major metropolitan centers (Dallas–Fort Worth, the Texas Coast including Houston, and the Interstate-35 corridor including Austin). Depending upon the season, these metroplexes can impact regional aerosol concentrations and radiative forcing up through the Central Plains. Two major air quality studies have been conducted in the gulf coast region of Texas: the Texas Air Quality Study 2000 (TexAQS 2000) and the Texas Air Quality Study 2006 (TexAQS 2006). These studies indicate high potential for urban outflow of organic matter and secondary aerosol from Texas metropolitan areas (Fan et al., 2005; Bates et al., 2008; Bahreini et al., 2009; Massoli et al., 2009; Russell et al., 2009; Zhang and Ying, 2010). A more recent study characterizing $PM_{2.5}$ in Houston, TX, identifies industrial activities and traffic in and around the Houston Ship Channel as major sources of $PM_{2.5}$ (Sullivan et al., 2013). With this outflow of major urban areas combining with regional agricultural and biogenic sources, central Texas is a natural laboratory for characterizing EC and BrC from mixed urban/rural sources.

Measurement of organic carbon (OC), elemental carbon (EC), and water-soluble organic carbon (WSOC) provides a broad perspective on carbonaceous aerosol sources and variability (Birch and Cary, 1996; Currie et al., 2002; Schauer et al., 2003). WSOC can comprise 20–70% of carbonaceous aerosol (Kirillova et al., 2010). WSOC is emitted by primary sources (biomass burning), is produced in the atmosphere by gas-to-particle conversion processes (secondary organic aerosol formation), and can be used to track aging of particulate carbon (Hecobian et al., 2010; Zhang et al., 2012; Kirillova et al., 2013). Several campaigns have measured WSOC concentrations in the central United States; however, these campaigns do not include absorption parameters for WSOC (Anderson et al., 2008; Snyder et al., 2009; Asa-Awuku et al., 2011). This study is the first in the central United States to include long-term WSOC absorption measurements.

This study presents filter-based ambient concentrations and absorption of OC, EC, and WSOC combined with calculation of b_{abs} and MAC for both the filter samples and aqueous extract. Statistical analysis combined with clustering of HYSPLIT back trajectories (BTs) was used to understand seasonal and source region influence on ambient concentrations.

Materials and Methods

Sampling site

A 15-month-long sampling campaign was conducted at the U.S. Department of Agriculture–Agricultural Research Service (USDA-ARS) Grassland, Soil, and Water Research Laboratory's Riesel Watersheds, Riesel, TX (31°28'30" N, 96°55'64" W). USDA-ARS Riesel Watersheds is a part of the Agriculture Research Service and managed by the U.S. Department of Agriculture and has been in operation since the mid-1930s (Harmel et al., 2003; Harmel et al., 2006; Harmel et al., 2007). Riesel is 29 km southeast of Waco, TX (Figure 1). The primary local sources are agricultural activities. The USDA-ARS site is located approximately 4 km off Texas Highway 6, the primary route between Waco and Houston, with annual average daily traffic count of 12,000 vehicles (2011, http://www.txdot.gov/inside-txdot/division/transportation-planning/maps/traffic.html).

Sample collection

$PM_{2.5}$ samples were collected once every six days from May 2011 through August 2012, following the U.S. Environmental Protection Agency (U.S. EPA) 1-in-6 day monitoring schedule, on a URG 3000b medium-volume sampler (URG Corporation, Research Triangle Park, NC). Teflo™ Teflon Membrane Disc Filters (47 mm) and quartz fiber filters (QFF, 90 mm) (Pall Corporation, Ann Arbor, MI) were used for sample collection. The sampler operated for 24 hr at 92 L min^{-1}, split at 10 L min^{-1} for the 47-mm filters and 82 L min^{-1} for the 90-mm filters, with a total of 132.48 m^3 of air sampled from midnight to midnight each day. Prior to sampling, the QFF were baked at 500°C for 12 hr and were individually stored in petri dishes lined with baked aluminum foil. All samples were stored in a freezer prior to and post sampling. Blanks were handled in the same manner as the sampled filters. Summer and winter intensives were conducted in August 2011 and January 2012. Twenty-four-hour samples were taken every other day over a 2-week period.

$PM_{2.5}$ data from Houston (Clinton St.), Dallas (Hinton St.), and Waco (Mazanec) monitoring stations was accessed via the Texas Commission on Environmental Quality (TCEQ) air quality monitoring website (http://www.tceq.texas.gov/cgi-bin/compliance/monops/site_photo.pl). Houston Clinton and Dallas Hinton OC and EC data were downloaded via the EPA Air Quality systems data mart available at http://www.epa.gov/ttn/airs/aqsdatamart/access/interface.htm (2012).

Bulk carbon and $PM_{2.5}$ analysis for Riesel

OC and EC concentrations were determined on a thermo-optical transmission (TOT) carbon analyzer (Sunset Laboratories, Tigard, OR) using the NIOSH 5040 method (Birch and Cary, 1996). An instrument blank and sucrose standard were run with every batch of 10 samples. All samples were blank subtracted using an average blank value of 0.19 μg cm^{-2}. For use in $PM_{2.5}$ mass balance, the OC concentrations need to be converted to organic mass (OM). This factor is dependent upon emission source composition and photochemical aging. Different factors have been recommended in the literature; for

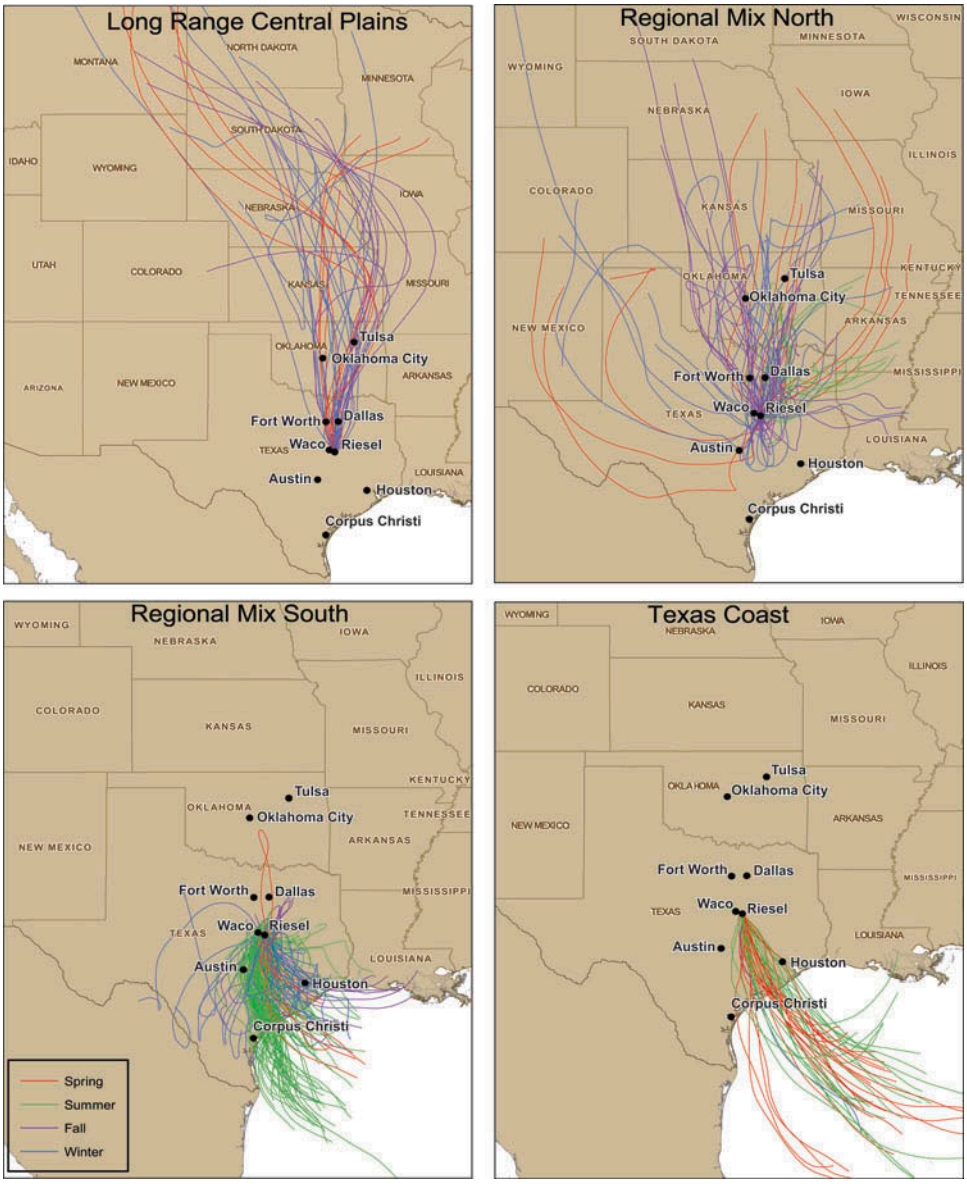

Figure 1. Forty-eight-hour back-trajectory plots showing the path of air arriving at Riesel, TX. Spring is shown in red, summer in green, fall in purple, and winter in blue.

example, a factor of 2.1 ± 0.2 is recommended for aged aerosols at rural sites downwind of urban areas (Polidori et al., 2008). In this case, an OM:OC factor determined by Fourier-transform infrared (FTIR) fluorescence during the TexAQS 2006 study in Houston (Russell et al., 2009) was used. A factor of 1.8 ± 0.14 was estimated for northerly air masses impacting Houston, and this factor is uniformly applied to estimate OM for Riesel (Russell et al., 2009). This factor is potentially biased high during winter seasons, which have less photochemical activity, and biased low for aged air masses impacting Riesel during summer months.

Prior to April 2009, the reported OC and EC values from the Dallas Hinton site were determined using the NIOSH protocol. The NIOSH protocol was also used at the Houston Clinton site prior to January 2010. After these dates, OC and EC concentrations for the Houston Clinton and Dallas Hinton sites were determined using the IMPROVE method. The two methods have been shown to report equivalent total carbon concentrations; however, EC concentrations from the NIOSH method have been shown to be lower than those from the IMPROVE method, depending on emission sources (Chow et al., 2001; Wright et al., 2010; Zhang et al., 2013). In order to address the possible bias in EC concentrations due to differences between the NIOSH and IMPROVE protocols, long-term trends in the EC and OC averages at the two urban sites were examined (Supplemental Materials, Figures S1 and S2). One-in-six 24-hr EC and OC concentrations from January 2006 through March 2009 were compared to concentrations from April 2009 to 2012 for the Dallas Hinton site, while measurements from 2006 through 2009 were compared to those for 2010 and 2011 for the Houston

Clinton site using an unpaired *t*-test ($\alpha = .05$). No significant differences in the EC concentrations from the NIOSH and IMPROVE methods were found using the statistical analysis at either the Houston Clinton (P-value = 0.06) or the Dallas Hinton (P-value = 0.08) site.

$PM_{2.5}$ mass was determined gravimetrically using the 47-mm Teflon filters. All filters were weighed in a humidity- and temperature-controlled room at the Wisconsin State Laboratory of Hygiene prior to and post sampling. $PM_{2.5}$ mass for Riesel was compared to data collected by the TCEQ at the Waco (Mazanec) air quality monitoring station ($r^2 = .55$). The Waco (Mazanec), Houston Clinton, and Dallas Hinton monitoring sites determine $PM_{2.5}$ mass using a tapered-element oscillating microbalance (TEOM). The TEOM has been shown to underestimate $PM_{2.5}$ mass when compared to gravimetrical measurements (Li et al., 2012), and this may explain the positive bias in the comparison between Riesel and Waco (Supplemental Materials, Figure S3). Despite the possible underestimation by the TEOM, $PM_{2.5}$ concentrations in Dallas and Houston are, on average, greater than Riesel and Waco $PM_{2.5}$ concentrations.

WSOC analysis

Water-soluble organic carbon (WSOC) analysis was performed on subsamples of the QFFs. WSOC extraction methods largely followed established protocols (specific extraction techniques are further outlined in the supplemental materials) (Kirillova et al., 2010; Ram and Sarin, 2010). Briefly, soluble compounds on the QFF were extracted by sonication using 30 mL of deionized water. Blank levels were on average 6% of the sample. WSOC concentrations were measured as dissolved organic carbon in the solution using a Shimadzu total organic carbon (TOC) analyzer (model TOC-5000A, Shimadzu, Kyoto, Japan) (Yang et al., 2003) (Supplemental Material, section S1).

Absorption parameters

Elemental carbon absorption. The optical attenuation (Wood et al., 2010) using the 678-nm laser source in the TOT analyzer was used to determine b_{abs} and MAC as described by Ram and Sarin (2009) and Cheng et al. (2011). All MAC calculations in this study have been corrected by an empirical correction factor, $C = 3.6$, used to correct the measured absorption for multiple scattering and shadowing effects (Weingartner et al., 2003; Ram and Sarin, 2009) (Supplemental Materials, section S3.1).

Brown carbon absorption. Light absorption of the WSOC extracts was measured at 200 to 700 nm on an Agilent 8453 ultraviolet–visible (UV-Vis) spectrometer (Santa Clara, CA), with deuterium and tungsten halogen light sources. Absorption coefficients were calculated at 365 nm (MAE_{365}) in order to avoid possible interferences from inorganic compounds, and to remain consistent with previously published BrC absorption parameters (Hecobian et al., 2010; Cheng et al., 2011). Further details on absorption calculations for BC and BrC are included in the Supplemental Materials (section S3).

Back trajectory and seasonal analysis. Forty-eight-hour back trajectories (BTs) were calculated for Riesel every 6 hrs for each sampling day over the entire field campaign in order to determine the geographic source region of air masses impacting the site. BTs were completed using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, version 4, May 2012 release (Draxler and Rolph, 2010), and were clustered using HYSPLIT. The HYSPLIT model was run using meteorological data from the Global Data Assimilation System (GDAS) produced by the National Oceanic and Atmospheric Administration. Each back trajectory was set to run with a starting height of 10 m above ground level, using vertical velocity fields supplied with the GDAS meteorological data. The resulting clusters were mapped using ESRI's ArcGIS 10 software on a map of Texas counties provided by ESRI (Figure 1). There are intrinsic uncertainties characteristic of the HYSPLIT model concerning modeling of terrain height; however, uncertainties of the current application are minimized due to the short duration of the back trajectories (48 hr) and the flat terrain of the region.

The field campaign was divided into four seasons (spring, summer, fall, and winter) based on examination of daily maximum temperatures, rather than a traditional three-month division, in order to capture the effect of high photochemistry in the late summer months in Texas. Each day was assigned to a season based on its maximum temperature to differentiate between seasons. Winter was defined as daily maximum temperatures of 0–15°C, spring 15–30°C, summer 30–42°C, and fall 30–15°C (blue, red, green, and purple, respectively, in Figure 1). Fall and spring have the same temperature range because they are transition seasons between the summer and winter and are distinguished by the time of year. This temperature range was chosen in order to capture the extended period of high photochemistry during the summer.

Results and Discussion

Seasonal analysis

Carbon and $PM_{2.5}$ mass measurements were grouped by time of year to assess effects of season on regional background aerosol concentrations in central Texas. Seasonal concentrations for the regional site are compared to values reported by TCEQ for Dallas, Houston, and Waco, TX.

Seasonal $PM_{2.5}$ and OC results. There are distinct seasonal trends for concentrations of $PM_{2.5}$ mass and OC in central Texas (Tables 1 and 2). Mean $PM_{2.5}$ concentrations for Riesel ranged from a springtime average of 6.8 ± 2.5 μg m^{-3} to a summertime average of 10.9 ± 4.7 μg m^{-3}. Studies of particulate matter in the southeastern United States have shown similar seasonal trends, with maximum $PM_{2.5}$ concentrations occurring in warmer months and the lowest concentrations for the cooler winter months (Chen et al., 2012; Zhang et al., 2012). This summertime peak is consistent with $PM_{2.5}$ monitoring stations in Waco and Dallas (Table 1). Houston peaks in spring, but has the second highest $PM_{2.5}$ in the summer months. The Houston Clinton monitoring site historically has one of the highest annual $PM_{2.5}$ concentrations in Texas; however, $PM_{2.5}$ concentrations have steadily declined since 2006 (Sullivan et al., 2013). Riesel differs

Table 1. Comparison of fine particulate matter ($PM_{2.5}$), organic carbon (OC), elemental carbon (EC), and EC/OC ratios for four sampling sites across Texas for 2011 and 2012

	Riesel (2011 and 2012)	Waco (2011 and 2012)	Houston Clinton (2011)				Dallas Hinton (2011 and 2012)			
Season	$PM_{2.5}$ ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	OC ($\mu g\ m^{-3}$)	EC ($\mu g\ m^{-3}$)	EC/OC ratio	$PM_{2.5}$ ($\mu g\ m^{-3}$)	OC ($\mu g\ m^{-3}$)	EC ($\mu g\ m^{-3}$)	EC/OC ratio
Spring (13)	6.77	9.78	14.1	3.73	0.67	0.19	11.9	2.26	0.42	0.19
Summer (39)	10.9	10.0	13.4	2.88	0.69	0.27	12.5	2.27	0.38	0.18
Fall (15)	8.50	7.43	10.4	3.58	0.78	0.23	10.6	2.00	0.51	0.25
Winter (18)	7.85	6.79	10.7	3.59	0.81	0.21	11.5	2.28	0.49	0.21

Note: The number of sampling days for each season is shown in parentheses next to each season.

from these three urban Texas sites in that springtime has the lowest $PM_{2.5}$ values. The Riesel results indicate important seasonal differences between urban and regional background $PM_{2.5}$.

The OC concentrations at Riesel ranged from $1.9 \pm 0.7\ \mu g\ m^{-3}$ in the spring to $3.0 \pm 1.6\ \mu g\ m^{-3}$ in the summer; however, the differences among the seasons were not statistically significant (*P*-values = 0.19, 0.21, 0.06, and 0.34 for winter, spring, summer, and fall, respectively). OC accounts for 27% of summer $PM_{2.5}$ mass (OM is 49%); this contribution is consistent for spring and summer at Riesel. OC concentrations in Dallas were consistent through winter, spring, and summer and decreased in the fall. Houston displays opposite trends for OC, with the lowest average value in the summer months and highs in the spring. Back-trajectory analysis can help to explain some of these regional differences; Houston in the summer has dominant southerly winds that have lower documented OC than northerly air masses due to fewer upwind anthropogenic sources (Bates et al., 2008). Absolute OC concentrations are very similar for Dallas and Riesel, while Houston is 40% higher on average (Tables 1 and 2).

For both Houston and Dallas sites, $PM_{2.5}$ concentrations have the highest correlation to OC and sulfate when compared to correlations with nitrate, ammonium or EC (Figures S4–S6 in Supplemental Materials). Regression analysis for Houston Clinton indicates roughly one-third of the variability in $PM_{2.5}$ concentrations can be explained by either sulfate or OC (r^2 = 0.32 and 0.39, respectively when regressed with $PM_{2.5}$). This correlation between sulfate and $PM_{2.5}$ is confirmed by Sullivan et al. (2013), who showed sulfate as one of the main contributors to $PM_{2.5}$ mass at Houston Clinton. For Dallas Hinton, sulfate and OC are also highly correlated with $PM_{2.5}$ concentrations (r^2 = 0.57 and 0.36, respectively, when regressed with $PM_{2.5}$). When summing the bulk inorganic and carbon components, 65% of the $PM_{2.5}$ variability can be explained at Texas urban sites ($r^2 = 0.65$ for sum of sulfate, nitrate, ammonium, OM, and EC when regressed with $PM_{2.5}$ at both Dallas and Houston sites) (Figures S4–S6). The TexAQS campaigns also documented significant contribution from sulfate to $PM_{2.5}$ in the Houston region (Bates et al., 2008), and related this to coal-fired electricity generation from upwind/northerly sources (Zhang and Ying, 2010).

The OC and $PM_{2.5}$ concentrations are correlated at Riesel on an annual scale with a correlation coefficient of 0.48, which is higher than either Dallas or Houston (Figure S7). On average, the OC accounts for 31% of the $PM_{2.5}$ mass at Riesel (OM accounts for 56%), with seasonal peaks in both summer and fall. Although sulfate measurements are not yet available for Riesel, it would be expected that regional sulfate would also be important to $PM_{2.5}$ concentrations at Riesel. Based on these differences in seasonal OC and $PM_{2.5}$ between Riesel and urban areas in Texas (Houston, Dallas, Waco), it is hypothesized that central Texas OC is impacted by a combination of transported urban and regional rural emissions.

Seasonal EC results. EC concentrations at Riesel are much lower than Dallas and Houston (factor of 2.4 and 4, respectively) (Tables 1 and 2). Despite this difference, EC at Riesel displays similar seasonal trends to Houston and Dallas, with fall/winter maximums and spring/summer minimums (Table 2). This is very

Table 2. Seasonal organic carbon (OC), elemental carbon (EC), organic mass (OM), EC/OC ratio, water-soluble organic carbon (WSOC), and WSOC/OC ratio concentrations for Riesel, TX, for the 15-month sampling campaign from May 2011 to August 2012

Season	OC ($\mu g\ m^{-3}$) ± SD	EC ($\mu g\ m^{-3}$) ± SD	OM ($\mu g\ m^{-3}$) ± SD	EC/OC ratio	WSOC ± SD	WSOC/OC ratio
Spring (13)	1.9 ± 0.73	0.17 ± 0.04	3.4 ± 0.74	0.09	1.3 ± 0.55	0.66
Summer (39)	3.0 ± 1.6	0.14 ± 0.07	5.3 ± 1.6	0.05	2.0 ± 1.3	0.66
Fall (15)	2.9 ± 2.4	0.22 ± 0.14	5.2 ± 2.4	0.08	1.9 ± 1.6	0.64
Winter (18)	2.3 ± 1.3	0.22 ± 0.09	4.2 ± 1.3	0.10	1.3 ± 0.76	0.55

Note: The number of sampling days for each season is shown in parentheses next to each season.

different from the $PM_{2.5}$ and OC trends discussed in the "Seasonal $PM_{2.5}$ & OC results" section, where OC at Riesel had different trends than urban centers but similar absolute concentrations. Dallas and Houston EC concentrations are typical of large urban areas throughout the United States (i.e., Pasadena, CA, and Atlanta, GA), while the Riesel concentrations are much lower (Zhang et al., 2011). Houston EC concentrations are generally higher than Dallas and Riesel for all seasons, potentially due to the large amount of diesel combustion sources in the Houston shipping channel in addition to urban traffic sources (Wu et al., 2009; Kuwayama et al., 2013). The Riesel EC/OC ratio was highest in the winter (0.10) and lowest in the summer (0.05). This difference could be driven by increased EC emissions during the winter months, possibly from residential wood burning or motor vehicle exhaust in the colder winter months. In addition to emission source changes, the back-trajectory analysis indicates a shift to dominant northerly air masses in the winter. These northerly air masses consistently pass over the Dallas–Fort Worth metroplex before arriving in Riesel. Based on this seasonal analysis, EC concentrations at Riesel are likely dominated by transport and dilution from urban areas, which may include areas with industrial as well as gas and oil production emissions, such as the Houston shipping channel. If EC is considered a tracer for urban impacts on Riesel, it further supports the notion that urban OC is supplemented by regional rural sources in central Texas.

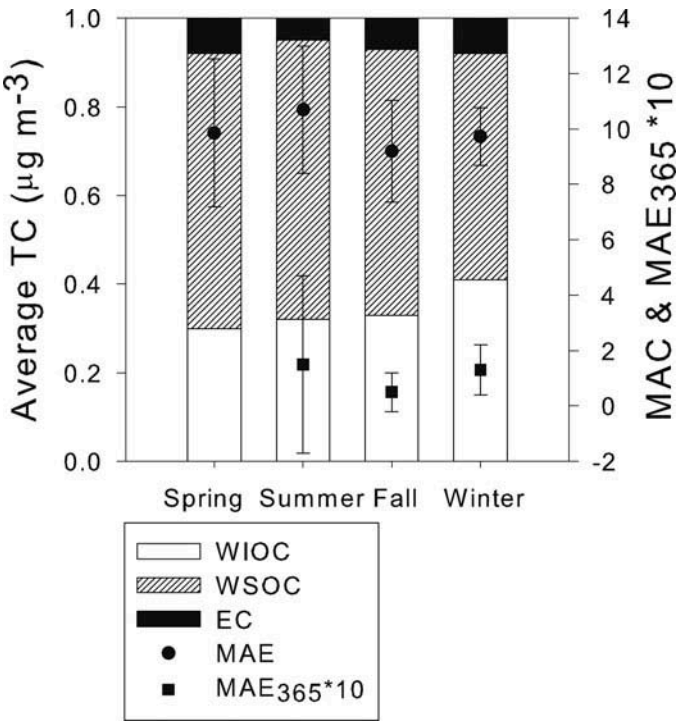

Figure 2. Contributions of water-insoluble organic carbon (WIOC), water-soluble organic carbon (WSOC), and elemental carbon by season at Riesel, TX. Average MAC and WSOC MAE_{365}*10 values are also presented. The WSOC MAE_{365} is not available for spring.

Seasonal WSOC results. To better understand the bulk carbon in central Texas, the water-soluble fraction was quantified. Mean WSOC concentrations ranged from 1.3 ± 0.55 $\mu g\ m^{-3}$ in the spring to 2.0 ± 1.30 $\mu g\ m^{-3}$ in the summer and showed similar seasonal trends as OC, with maximum concentrations in the summer and fall (Table 2 and Figure 2). However, the differences among seasons were not statistically significant (P-values = 0.10, 0.17, 0.31, and 0.08 for winter, spring, summer, and fall, respectively). Summer WSOC values for Riesel are higher than summer WSOC concentrations in Pasadena, CA (1.0 ± 0.72), and similar to summer WSOC concentrations in Atlanta, GA (2.0 ± 1.0), which has known high biogenic contributions (Zhang et al., 2011). The small increase in WSOC concentrations in the summer and fall are potentially due to an enhancement of secondary chemical processes due to increased solar radiation and biogenic VOC emissions (Chow et al., 2009; Zhang and Ying, 2011; Zhang et al., 2012). Photo-oxidation of nonmethane hydrocarbons (NMHC) from oil and gas production in the source regions could also contribute to the higher concentrations (Berkowitz et al., 2005; Carlton et al., 2010). However, at a rural site like Riesel, it is also likely that high WSOC is related to aging of OC during transport from source areas (Kirillova et al., 2013). WSOC/OC ratios were lowest in the winter months, at 0.55, and increased to 0.66 during the spring and summer (Table 2). Regression of WSOC with OC gives a correlation coefficient of $r^2 = 0.90$ with a slope of 0.70. Given the small difference among seasons and the consistent relationship to OC, WSOC is likely a mixture of aged urban and aged regional rural aerosol.

Back-trajectory cluster analysis

Forty-eight-hour BTs showed that air masses impacting the sampling site could be divided into four distinct clusters: the

Table 3. Seasonal averages of elemental carbon light attenuation (EC ATN), elemental carbon mass absorption efficiency (EC MAC), water-soluble organic carbon ATN (WSOC ATN), absorption coefficients of WSOC extracts(WSOC b_{abs}), and mass absorption efficiency at 365 nm (WSOC MAE_{365}) of WSOC extracts

Season	EC ATN ± SD	EC MAC ($m^2\ g^{-1}$) ± SD	WSOC ATN ± SD	WSOC $b_{abs}(Mm^{-1})$ ± SD	WSOC MAE_{365} ($m^2\ g^{-1}$)
Spring	0.22 ± 0.06	9.9 ± 2.7	N/A	N/A	N/A
Summer	0.21 ± 0.13	11 ± 2.3	0.013 ± 0.05	0.35 ± .11	0.15 ± 0.32
Fall	0.29 ± 0.16	9.2 ± 1.8	0.002 ± 0.002	0.08 ± 0.12	0.05 ± 0.07
Winter	0.32 ± 0.15	9.7 ± 1.0	0.004 ± 0.003	0.17 ± 0.14	0.13 ± 0.09

Note: Spring WSOC ATN, WSOC b_{abs}, and WSOC MAE_{365} values not available.

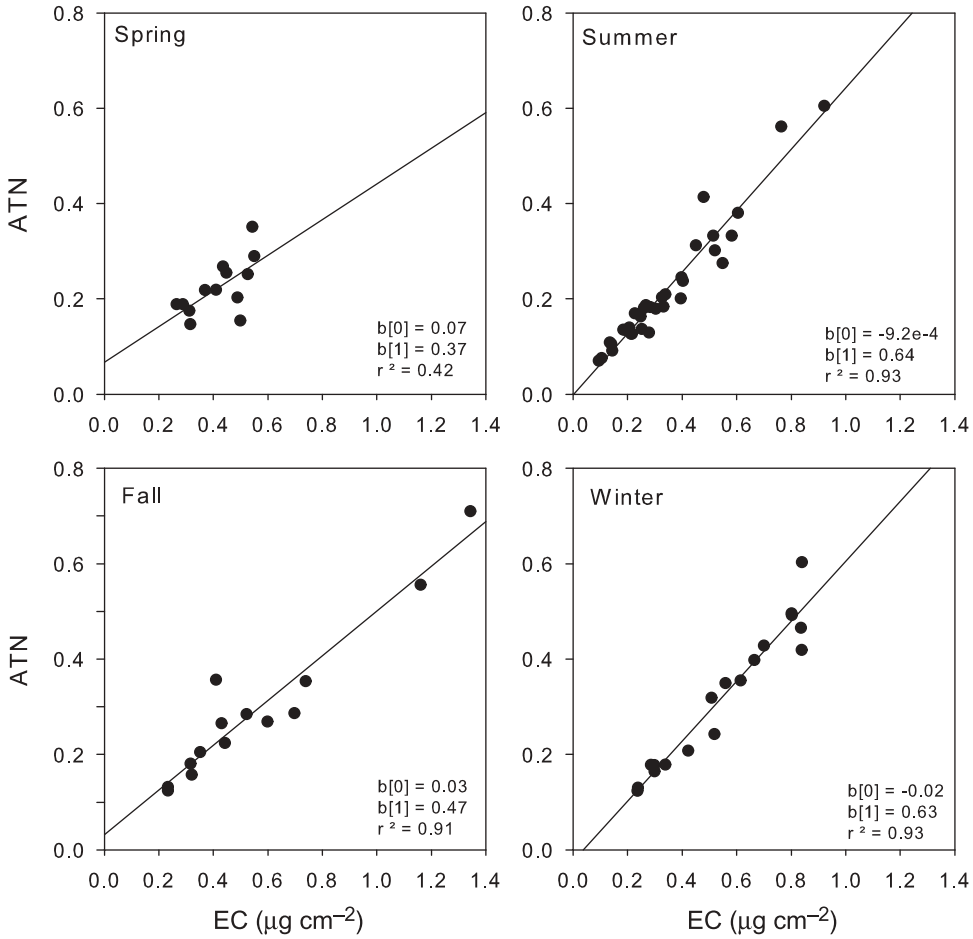

Figure 3. Light attenuation vs. filter elemental carbon (EC) loading by season; b[0] is equal to the *y*-intercept and b[1] is equal to the slope of the regression.

Long Range Central Plains cluster, the Regional Mix North cluster, the Texas Coast cluster, and the Regional Mix South cluster (Figure 1). This annual BT analysis indicates that the Central Plains and Central Texas can be considered to be connected airsheds. Geographic source analysis of contributions of OC, EC, and WSOC showed no significant difference by BT cluster with the exception of lower EC concentrations from air masses originating in the Regional Mix South Cluster (*P*-value = 0.002) (Supplemental Material, section S3 and Table S1). It has previously been shown that Houston, TX, a coastal city, exhibits a pronounced dependence on BTs for OC concentration (Bates et al., 2008); however, there is a higher dependence on season rather than source region in explaining the variability in carbon components in central Texas.

Absorption

Seasonal absorption. EC light attenuation (ATN) is consistent throughout the year, ranging from 0.21 ± 0.13 in the summer to 0.32 ± 0.15 in the winter, indicative of a steady source of dilute

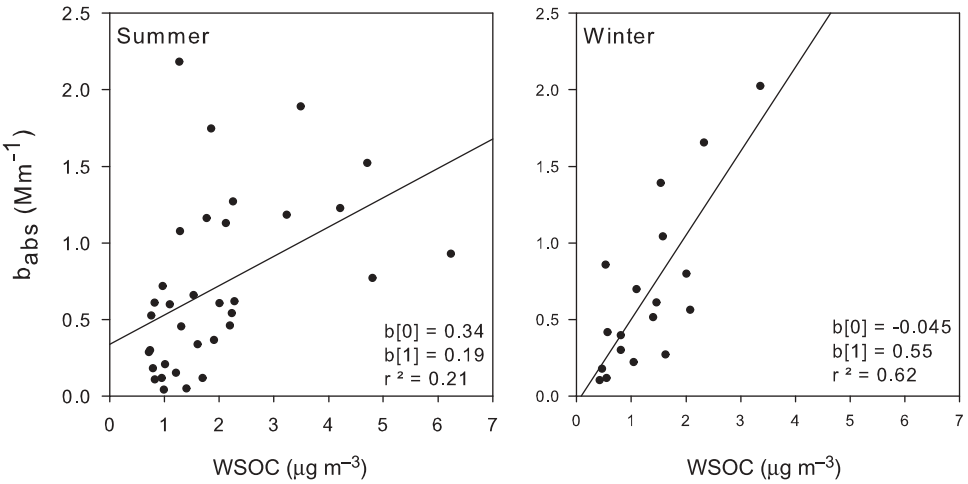

Figure 4. Scatter plots showing the correlation of WSOC b_{abs} (365 nm) and water-soluble organic carbon (WSOC) for summer and winter at Riesel, TX; b[0] is equal to the *y*-intercept and b[1] is equal to the slope of the regression.

urban EC (Table 3). ATN and EC filter loadings exhibit a strong correlation for all seasons other than spring (r^2= 0.9 for summer, fall and winter, r^2= 0.42 for spring, Figure 3), indicating that EC is the principal absorbing component for most of the year. The weak correlation in spring (r^2 = 0.42) indicates contribution of absorption from other species (possibly BrC or mineral dust). There is no significant difference in the slopes for summer, winter, and fall at the 95% confidence interval according to Student's *t*-test (n = 3). The MAC calculated at 678 nm ranged from 8.5 to 14.6 with a mean standard deviation of ±1.0 m^2 g^{-1} in summer. In winter the MAC ranged from 8.3 to 12.1 with a mean standard deviation of ±2.3 m^2 g^{-1}. It has been shown that MAC is influenced by mixing state, and when EC is internally mixed with components such as sulfate and OC, these coatings can focus light into the EC core, increasing the MAC (Cheng et al., 2011). Maximum MAC values coincide with maximum OC and WSOC concentrations during the summer (Figure 2), potentially indicating the intensification of MAC by organic aerosol coating, including secondary organic aerosol (SOA).

Seasonal BrC absorption. Light absorption of the WSOC aqueous solutions was explored in order to determine the presence and potential regional climate effects of BrC (Table 3). WSOC ATN is much higher in summer, 0.013 ± 0.05, than in fall and winter, 0.002 ± 0.002 and 0.004 ± 0.003, respectively, and is likely connected to summer photochemical events. The WSOC MAE_{365} averaged 0.15 ± 0.32 m^2 g^{-1} for summer and 0.13 ± 0.09 m^2 g^{-1} for winter (Figure 2). BrC has been shown to rapidly form in high NO_x (urban) environments, and nitrogen-containing aromatic compounds are effective light absorbers at 365 nm, with urban WSOC MAE_{365} values generally higher than those of biogenic SOA (Zhang et al., 2013). Average WSOC MAE_{365} in Riesel is much lower than the reported average value of 0.71 m^2 g^{-1} for summer 2010 in Los Angeles, CA, a high-NO_x environment (Zhang et al., 2013). Average WSOC b_{abs} values were 0.17 ± 0.14 and 0.35 ± 0.14 Mm^{-1} for winter and summer, respectively. The summer maximum seen in Riesel is lower than summer values reported for the Atlanta, GA (0.61 ± 0.38 Mm^{-1}), and Pasadena, CA (0.88 ± 0.71 Mm^{-1}), urban areas (Zhang et al., 2011). This difference may be due to a lower number of anthropogenic mobile sources and higher biogenic or NMHC sources of BrC at Riesel. WSOC and WSOC b_{abs} were positively correlated during winter (r^2= 0.62). The correlation was low during summer, but this may be due to variable WSOC:WSOC b_{abs} ratios during this time (Figure 4). The absorption spectrum of the WSOC extracts for both summer and winter are typical of BrC, with absorption decreasing sharply as wavelength increases (Figure 5) (Andreae and Gelencser, 2006; Hoffer et al., 2006; Cheng et al., 2011).

Conclusion

Absolute OC concentrations are in the same annual range for Riesel, Dallas, and Houston throughout the study period. Strong correlation of OC and $PM_{2.5}$ at Riesel and differing seasonal trends between Riesel and Texas urban areas supports the notion that dispersed rural OC sources contribute significantly to regional $PM_{2.5}$ concentrations in the Central USA. Based on the BT analysis, these rural sources of OC could impact baseline $PM_{2.5}$ concentrations for urban areas including Houston and Dallas–Fort Worth. WSOC:OC ratios remain consistent at Riesel regardless of season or source region. A consistent annual relationship of WSOC to OC points to the importance of aged atmospheric aerosols arriving at Riesel. OC reaching Riesel is likely a mixture of aged urban and aged regional rural aerosol.

Absolute EC concentrations in Dallas and Houston are four to seven times higher than in Riesel for the study period. The EC:OC ratio decreases from the urban sites (Dallas and Houston) relative to Riesel by a factor of 2. Low EC concentrations at Riesel are likely due to a dilution of urban EC during transport with minimal EC contributions from regional rural/agricultural sources as compared to OC.

Absorption due to both EC and BrC was explored in this study in order to provide a regional perspective on potential climate forcing by atmospheric aerosols. Biogenic SOA predominance, in addition to SOA from NMHC sources, may contribute to the lower WSOC MAE_{365} values as compared to urban

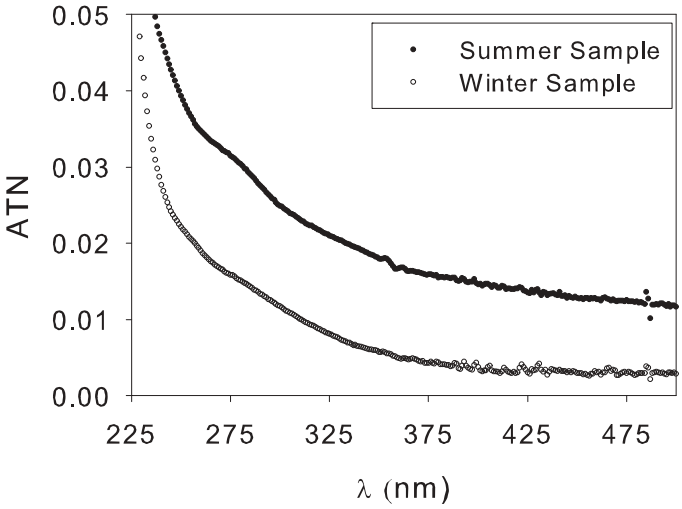

Figure 5. Example of water-soluble absorption spectra for one summer (08/13/2012) and one winter (02/15/2012) sample from Riesel, TX. The Angstrom absorption exponent of the summer sample was 5.8, and 4.8 for the winter sample.

sites (Zhang et al., 2013). These WSOC MAE_{365} results also support the notion of rural OC impacting Riesel. BrC ATN peaked during the summer. The combined optical results indicate the dominance of aerosol absorption by EC in central Texas. BrC does contribute to absorption in the summer at Riesel; however, the lower WSOC MAE_{365} indicates less absorption by unit mass for the WSOC in Riesel compared to the southeastern United States and California. Continued monitoring of this site will improve understanding of regional aerosol absorption in the central United States.

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Supplemental Data

Supplemental data for this article can be accessed on the publisher's website.

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