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TECHNICAL PAPER

The Southeastern Aerosol Research and Characterization (SEARCH) study: Spatial variations and chemical climatology, 1999–2010

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The Southeastern Aerosol Research and Characterization (SEARCH) study, which has been in continuous operation from 1999 to 2012, was implemented to investigate regional and urban air pollution in the southeastern United States. With complementary data from other networks, the SEARCH measurements provide key knowledge about long-term urban/nonurban pollution contrasts and regional climatology affecting inland locations and sites along the Gulf of Mexico coastline. Analytical approaches ranging from comparisons of mean concentrations to the application of air mass trajectories and principal component analysis provide insight into local and area-wide pollution. Gases (carbon monoxide, sulfur dioxide, nitrogen oxides, ozone, and ammonia), fine particle mass concentration, and fine particle species concentrations (including sulfate, elementary carbon, and organic carbon) are affected by a combination of regional conditions and local emission sources. Urban concentrations in excess of regional baselines and intraurban variations of concentrations depend on source proximity, topography, and local meteorological processes. Regional-scale pollution events (95th percentile concentrations) involving more than 6 of the 8 SEARCH sites are rare (< 2% of days), while subregional events affecting 4–6 sites occur on ~10% of days. Regional and subregional events are characterized by widely coincident elevated concentrations of ozone, sulfate, and particulate organic carbon, driven by persistent synoptic-scale air mass stagnation and higher temperatures that favor formation of secondary species, mainly in the summer months. The meteorological conditions associated with regional stagnation do not favor long-range transport of polluted air masses during episodes. Regional and subregional pollution events frequently terminate with southward and eastward penetration of frontal systems, which may initially reduce air pollutant concentrations more inland than along the Gulf Coast.

Implications: Regional distribution of emission sources and synoptic-scale meteorological influences favoring stagnation lead to high regionwide pollution levels. The regional influence is greatest with secondary species, including ozone (O_3) particulate sulfate (SO_4), and particulate organic matter, some of which is produced by atmospheric oxidation of volatile organic compounds (VOCs) from vegetation and anthropogenic sources. Other species, many of which are from primary emissions, are more influenced by local sources, especially within the Atlanta, GA, and Birmingham, AL, metropolitan areas. Limited measurements of modern and fossil total carbon point to the importance of biological and biogenic emissions in the Southeast.

Introduction

Air pollutant observations across the United States reveal pollutant-specific concentration heterogeneities within urban areas and across nonurban regional surroundings (e.g., Magliano et al., 1999; Blanchard et al., 1999; Butler et al., 2003; Jerrett et al., 2005; Kanaroglou et al., 2005; Ott et al., 2008; Demerjian and Mohnen, 2008; Turner and Allen, 2008). These heterogeneities have implications for human population exposure assessment, epidemiological study of air pollution effects, and air quality management practices.

The southeastern United States represents unique climatologic and demographic conditions in a subtropical vegetative environment. The region is known to experience active atmospheric photochemistry, stimulated in part by the presence of biogenic organic compounds in addition to anthropogenic emissions, and with intense haze partly associated with persistent high humidity. Growth in population and industry is reflected in large, relatively isolated cities, such as Atlanta, GA, and Birmingham, AL, which experience elevated concentrations of a number of pollutants driven by local and regional contributions. Air pollution conditions reported in the region approach or exceed applicable air quality standards (U.S. EPA, 2012a), so that in-depth knowledge of the chemical climatology of the area is important for management of air quality.

Investigation of multiscale spatial distributions of pollutants in the Southeast is facilitated by the presence of a major, long-term program, the Southeastern Aerosol Research and Characterization (SEARCH) study, which has operated continuously from 1999 through 2012. This network of eight sites includes measurements

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of both gaseous and particulate species along with meteorological data, and complements monitoring by the federal and state governments and by other research studies. The SEARCH network is comprised of paired urban and nonurban sites, including sites in two major cities in the region, Atlanta, GA, and Birmingham, AL, two nonurban inland sites, and four sites located near the coast of the Gulf of Mexico. The project has been described in several papers, including Hansen et al. (2003) and Edgerton et al. (2005, 2006, 2007, 2009).

The configuration of the SEARCH network supports assessment of regional and local air pollution events, including their duration, extent, contributing emission sources, and meteorological drivers, in a large portion of the Southeast. Improved knowledge of the spatial variations of particulate matter (PM) and gas-phase pollutants within urban areas and across the Southeast in turn supports air pollution exposure assessment, epidemiological analysis, and source attribution. This paper evaluates air-pollutant compositional similarities and differences extending to a range of spatial scales for the different geographical settings of the SEARCH sites, based on more than 10 years of data from the SEARCH network. The spatial scales considered include a regional domain with linear dimensions of ~400 km N–S by ~600 km W–E, and an urban scale exemplified by the Atlanta and Birmingham metropolitan areas and their immediate surroundings (linear scale ~50–100 km). A companion paper is devoted to the assessment of temporal trends from 1999 to 2010 (Blanchard et al., 2013).

The 1999–2010 observations of carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NO_x and NO_y), reduced nitrogen (ammonia, NH_3; ammonium, NH_4), ozone (O_3), and $PM_{2.5}$ and $PM_{10\text{-}2.5}$ mass and species concentrations are summarized. Urban/nonurban contrasts and contrasts between inland locations and sites on or near the Gulf of Mexico coastline are discussed. Regional variations are evaluated using mean concentrations and principal component analysis of daily data. The intraurban conditions in Atlanta and Birmingham are examined using the SEARCH data along with additional gas and PM concentration measurements acquired by other monitoring programs.

Measurements

Network description

The SEARCH network was designed to provide long-term, detailed gas and particle chemistry for paired urban and nonurban sites in the Southeastern United States. Its design deemphasizes high-resolution characterization of pollutant gradients across the Southeast in favor of detailed long-term chemical knowledge at key locations. The eight stations provide insight into regional and urban pollution characteristics, especially when combined with data from other monitoring programs. SEARCH data are accessible from the SEARCH public archives (Atmospheric Research and Analysis, 2010).

The locations and detailed descriptions of the eight SEARCH sites (Figure 1) are discussed in Hansen et al. (2003). Pensacola (PNS) and Gulfport (GFP) are urban sites located ~5 km and 1.5 km from the Gulf Coast shoreline, respectively; Pensacola—outlying (aircraft) landing field (OLF) and Oak Grove (OAK) are nonurban sites located ~15 km and 75 km from PNS and GFP, respectively. Atlanta—Jefferson Street (JST) and North Birmingham (BHM) are urban inland sites; Yorkville (YRK) and Centreville (CTR) are nonurban inland sites. OAK is least affected by neighboring urban exposure and is considered a regional baseline indicator at the western side of the domain. CTR is a rural, regionally representative site that experiences intermittent exposure from Birmingham, Montgomery, AL, and Tuscaloosa, AL; YRK is a nonurban site that is at times directly influenced by the Atlanta plume or by nonurban electric generating units (EGUs) located at distances of ~25 to ~85 km from YRK. JST is situated in midtown Atlanta, the largest city in the region, and is locally influenced by transportation and light industry. The BHM site is located in an industrial–residential area affected by a neighboring steel pipe plant (< 1 km distant), nearby coking operations (~5 km distant), motor vehicle and rail transportation, and more distant industrial and commercial operations. PNS and GFP are modestly populated commercial-resort communities along the Gulf of Mexico, with fewer major stationary sources but a significant influence from motor vehicle transportation. The PNS and OAK sites were closed in 2010 and 2011, respectively.

The SEARCH network operations, sampling, and measurement methods for gases (CO, SO_2, NO, NO_y, HNO_3, O_3, and NH_3) and fine ($PM_{2.5}$) and coarse ($PM_{10\text{-}2.5}$) particles are documented in Hansen et al. (2003, 2006), Edgerton et al. (2005, 2006, 2007, 2009), and Saylor et al. (2010). All sites have measurements of meteorological parameters and gas-phase species (i.e., criteria pollutants; HNO_3 and 24-hr NH_3 commencing mid-2003) at 10 m height (Hansen et al., 2003; Edgerton et al., 2007; Saylor et al., 2010). Measurements of speciated nonmethane organic compounds (NMOC) were made at JST through 2008 (Blanchard et al., 2010). Particulate measurements include 24-hr resolution filter-based or continuous $PM_{2.5}$ and $PM_{10\text{-}2.5}$ mass concentration and bulk composition of inorganic species (water-soluble ions and elements by x-ray fluorescence) plus organic carbon (OC) and elemental carbon (EC) (Hansen et al., 2003; Edgerton et al., 2005, 2006, 2009). SEARCH reports organic mass (OM) by scaling OC by a constant factor of 1.4 ($OM = 1.4 \times OC$), while recognizing that the actual scaling factor is not constant and may be higher on average (Hansen et al., 2003; El-Zanan et al., 2009). A second SEARCH convention reports concentrations of major metal oxides (MMO) as the sum of the highest oxidation states of six elements (i.e., $MMO = Al_2O_3 + SiO_2 + K_2O + CaO + TiO_2 + Fe_2O_3$). Although these metals may exist in different valence states, their oxidation states are specified here by the MMO convention. The concentrations of MMO, its component elements, and other elements are reported in the SEARCH database.

Carbon isotope (^{12}C and ^{14}C) measurements were made on daily-average Carbonaceous Aerosol Characterization Experiment (CACHE) samples collected from January 2004 through July 2005 at JST ($n = 48$) and from January through December of 2004 at BHM and CTR ($n = 46$) and PNS ($n = 39$) (Zheng et al., 2006; Blanchard et al., 2011). Samples for carbon isotope analysis were also collected in 2001–2002 at OAK ($n = 18$), YRK ($n = 19$), and JST ($n = 27$) over 3-day sampling intervals. The carbon isotope measurements partition total carbon ($TC = EC + OC$) into carbon derived from modern emissions (having higher ^{14}C) and carbon derived from combustion of fossil fuels (depleted in ^{14}C). The CACHE results are reported as the TC modern fraction and were

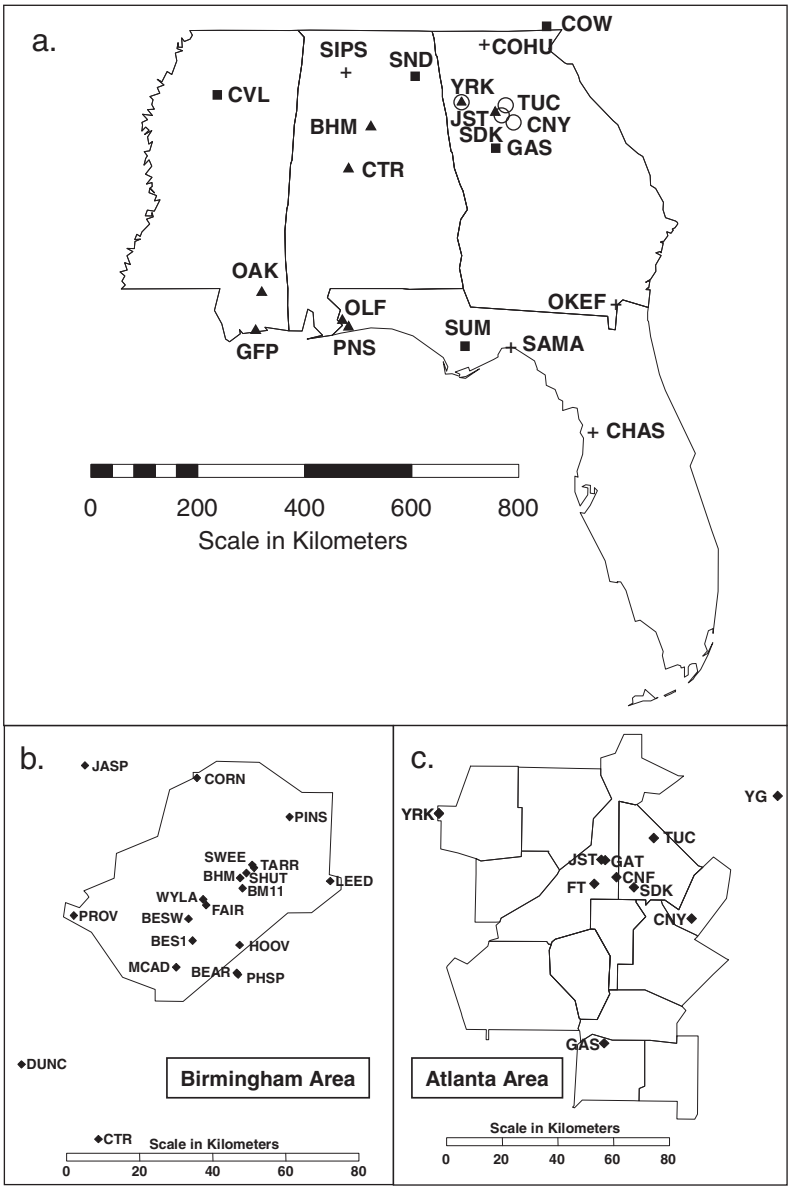

Figure 1. Location maps: (a) SEARCH (triangles), CASTNet (squares), PAMS (circles), and IMPROVE (plus symbols) monitoring sites; (b) SEARCH and additional monitoring sites in and near Birmingham with Jefferson County boundaries shown; (c) SEARCH and additional monitoring sites in and near Atlanta with county boundaries shown.

corrected for enhanced ^{14}C formation attributable to nuclear weapons testing using a 6% adjustment factor. Same-day CACHE and SEARCH samples were paired for calculating modern and fossil fractions of SEARCH TC.

To enhance area coverage, complementary aerometric observations were obtained from U.S. Environmental Protection Agency (EPA) compliance monitors and Photochemical Assessment Stations (PAMS) (U.S. EPA, 2009), Chemical Speciation Network (CSN) sites (U.S. EPA, 2009), and Clean Air Status and Trends Network (CASTNet) sites (U.S. EPA, 2010). PM data from two sites in the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, Sipsey (SIP), AL, and Cohutta (COH), GA were obtained from IMPROVE archives (IMPROVE, 2010). Daily PM mass and species concentrations have been made at four locations in the Atlanta area under the Assessment of Spatial Aerosol Composition in Atlanta (ASACA) program (Butler, 2003; ASACA, 2012), and current summary data were obtained from Trail (2010). The complementary data provide more spatial detail for the SEARCH domain, especially within the Birmingham and Atlanta metropolitan areas (Figure 1).

Supplementary meteorological data were obtained from the U.S. EPA Omnibus Meteorological Data (OMD) data set for the period 1999–2006 (U.S. EPA, 2008). The OMD includes two quantities (transport distance and transport direction) that are derived from air mass back trajectories computed using the HYSPLIT model (Draxler et al., 2009). Transport distance and direction are determined from the locations of back trajectories 24 hr prior to times of initiation relative to specified starting points. Trajectories are initiated each day at 12 noon local time at a height of 300 m above ground level (U.S. EPA, 2008). The

HYSPLIT model was used here to reproduce OMD transport distance and direction for a comparison time period, and to determine additional back trajectories for each day of 2007 through 2010. The full set of transport direction and distance data for the period 1999–2010 used starting locations at airports in Atlanta (n = 4254 days), Birmingham (n = 4347 days), and Mobile (n = 4254 days).

Methods of analysis

The regional spatial characteristics of gas and particle chemistry across the SEARCH domain and in the two larger cities, Birmingham and Atlanta, were investigated using the following approaches: (1) comparison of mean concentrations obtained over 12 years and shorter periods; (2) comparison of SEARCH and complementary data for evidence of spatial variations across the region and within the two largest cities; (3) spatial covariance assessment using principal component analysis; and (4) regional assessment using meteorological data and air mass trajectory characteristics to seek evidence of long range transport compared with air mass stagnation.

Results

Mean concentrations

Table 1 summarizes the long-term mean concentrations of gases and particles in the SEARCH regime from 1999 through 2010. Among the nonurban sites, 1999–2010 mean concentrations of the gas-phase species CO, NO, and NO_y were lowest at OAK, followed by CTR, YRK, and OLF in increasing order (Table 1). The nonurban mean SO_2 concentrations followed the same order as the CO, NO, and NO_y means, except that mean SO_2 concentration was greater at YRK than at OLF. OLF is a suburban site and may be more influenced by local emissions of CO and NO than are the rural sites at OAK, CTR, and YRK. The mean concentrations of CO, NO, NO_y, and SO_2 were ~2–5 times higher at BHM and JST than at GFP and PNS. The means of nonurban peak 8-hr O_3 concentrations indicate a regional value of 33–37 ppbv. In contrast, urban sites show lower mean peak 8-hr O_3 concentrations of ~25–29 ppbv. Since higher average levels of NO occur in urban than in nonurban areas, titration of O_3 by NO may lower urban peak O_3 concentrations relative to nonurban values.

Table 1. Mean concentrations by site determined from all data available for 1999–2010. $PM_{2.5}$ and $PM_{10\text{-}2.5}$ mass and species concentrations are means of daily measurements (24-hr filter-based samples), 1999–2010. NH_3 data are means of daily measurements (24-hr filter-based samples), 2004–2010. O_3 measurements are means of peak daily 8-hr maxima, 1999–2010. Concentrations of other gases are means of all hours, 1999–2010 (5–12 yr available data for NO_2 and 2–6 yr for HNO_3). Means were determined by year, and the yearly means were then averaged

	Coastal sites				Inland sites			
	Nonurban		Urban		Nonurban		Urban	
Species	OAK[a]	OLF	GFP[a]	PNS[b]	CTR	YRK	BHM[c]	JST
$PM_{2.5}$ ($\mu g\ m^{-3}$)								
Mass	11.06	10.69	10.93	12.28	11.51	12.89	16.53	15.46
SO_4	3.26	3.22	3.28	3.42	3.50	4.11	4.31	4.27
NO_3	0.32	0.36	0.42	0.42	0.33	0.76	0.83	0.87
NH_4	0.98	1.04	1.06	1.12	1.12	1.62	1.55	1.67
EC	0.50	0.60	0.62	0.78	0.58	0.57	1.76	1.34
OM	3.71	3.41	3.35	3.96	4.22	4.31	6.17	5.88
MMO	0.49	0.44	0.50	0.59	0.38	0.32	1.02	0.51
$PM_{10\text{-}2.5}$ ($\mu g\ m^{-3}$)								
Mass	6.10	6.59	8.08	9.06	5.25	5.23	14.38	8.28
SO_4	0.21	0.26	0.30	0.29	0.19	0.21	0.34	0.24
NO_3	0.45	0.56	0.72	0.64	0.37	0.37	0.48	0.41
NH_4	0.03	0.03	0.03	0.03	0.04	0.05	0.02	0.03
MMO	1.58	1.41	2.02	2.39	1.33	1.34	4.91	3.03
Gases (ppbv)								
O_3	34.81	32.95	32.54	29.12	35.18	37.68	24.66	24.47
CO	160.39	181.95	218.21	262.11	165.64	178.73	397.67	417.25
SO_2	1.29	1.76	1.62	2.30	1.66	2.31	4.20	4.33
NO	0.15	0.86	2.24	3.59	0.29	0.46	13.52	21.21
NO_2	1.36	4.22	5.68	7.29	2.23	3.8	15.34	18.84
HNO_3	0.39	0.32	0.31	0.23	0.62	0.86	0.54	1.03
NO_y	2.64	6.49	9.76	11.72	4.00	6.12	30.66	42.75
NH_3[d]	0.31	0.47	0.76	0.82	0.32	2.67	2.53	1.40

Notes: [a]$PM_{10\text{-}2.5}$ measurements at GFP and OAK start in 2003. [b]All PNS measurements end after 2009. [c]Gas measurements at BHM began in 2000. [d]NH_3 measurements commenced in 2004.

Table 1 indicates that NH_3 concentrations vary by nearly an order of magnitude across the domain, with lowest values at OAK and highest values at YRK and in the two major cities. Mean NH_3 concentrations range from 32% to 73% of the total reduced nitrogen measured. The nitrate data show total nitrate (HNO_3 plus fine and coarse particle NO_3) in the range of 1.7–4.2 μg m^{-3}, with higher values inland or in urban areas. Nitric acid ranges from 38% to 71% of total NO_3; coarse particle NO_3 mean concentrations tend to be higher than fine particle means, except in the inland urban areas and at YRK. Combining the gas and particle NO_y data (as NO_2), total NO_3 ranges from 4 to 29% of NO_y, with the highest fractions at CTR, YRK, and OAK. This result suggests regional aging of nitrogen oxides to increase the fraction of more highly oxidized components of NO_y relative to NO and NO_2.

Mean 1999–2010 $PM_{2.5}$ mass concentrations ranged from 10.7 to 12.3 μg m^{-3} at the coastal SEARCH sites and from 11.5 to 16.5 μg m^{-3} at inland SEARCH sites (Table 1). The $PM_{2.5}$ mass concentrations comprised 4.6–6.8 μg m^{-3} inorganic species (40–50% of $PM_{2.5}$ mass), 4.0–7.9 μg m^{-3} carbonaceous material (36–48% of $PM_{2.5}$ mass), and 0.3–1.0 μg m^{-3} MMO (2.6–6.2% of $PM_{2.5}$ mass) (Table 1).

The 1999–2010 mean $PM_{2.5}$ nitrate (NO_3) concentrations were 0.3–0.9 μg m^{-3} (2.9–5.6% of $PM_{2.5}$ mass) (Table 1). Mean particulate NO_3 concentrations were 0.3–0.4 μg m^{-3} at CTR and the coastal sites, and 0.8–0.9 μg m^{-3} at BHM, YRK, and JST (Table 1). Thermal equilibrium model applications indicate that $PM_{2.5}$ NO_3 formation in the Southeast is limited by the availability of NH_3 (Blanchard and Hidy, 2003, 2005; Blanchard et al., 2007). The mean coarse NO_3 concentrations exceeded mean fine NO_3 concentrations at all four coastal sites (Table 1), speculatively consistent with a known mechanism in which coarse NO_3 may be linked with displacement of chloride (Cl) from sea salt by nitric acid, forming $NaNO_3$. The inland coarse nitrate concentrations are on average ~30% lower than those found along the Gulf Coast. The origin of the inland coarse NO_3 component is uncertain, but hypothetical mechanisms include adsorption of HNO_3 on soil dust in analogy to the HNO_3–NaCl reaction or formation of $NaNO_3$ from lower inland concentrations of sea salt aerosol.

Coarse particle ($PM_{10\text{-}2.5}$) concentrations at rural sites (5.2–6.6 μg m^{-3}) are on the order of half of the fine particle concentrations, but are a much larger fraction in the urban areas, ranging from 57 to 90%. The coarse fraction contains substantial MMO and smaller amounts of SO_4, NO_3, and NH_4. Coarse SO_4 concentrations averaged 0.2–0.3 μg m^{-3}, less than 10% of fine SO_4 concentrations at all sites. The mean coarse NH_4 concentrations were less than 0.1 μg m^{-3}, which is insufficient to neutralize either the coarse SO_4 or the coarse NO_3. Alkaline metal cations possibly neutralize coarse SO_4 and coarse NO_3. For example, mean concentrations of coarse calcium (Ca) ranged from 0.17 to 0.45 μg m^{-3} (1.2 μg m^{-3} at BHM), sufficient to neutralize 0.3–0.8 μg m^{-3} coarse SO_4 (2.9 μg m^{-3} at BHM).

Coarse OC and EC measurements were made on subsets of samples at CTR (2000–2001, $n = 91$), YRK (2002–2004, $n = 127$), JST (2003–2004, $n = 242$), BHM (2008–2009, $n = 179$), and OLF (2008–2009, $n = 112$) (Edgerton et al., 2009). When paired with contemporaneous, co-located fine PM OC and EC measurements, mean coarse OC concentrations (1.2–2.2 μg m^{-3}) were less than mean fine OC (2.7–4.4 μg m^{-3}) concentrations. Also, mean coarse EC concentrations (0.1–0.3 μg m^{-3}) were less than mean fine EC concentrations (0.6–1.5 μg m^{-3}). At CTR, the mean coarse OC/EC ratio of 4.5:1 was essentially the same as the mean fine OC/EC ratio of 4.7:1. At JST and YRK, the mean coarse OC/EC ratios (9.1:1 and 10.5:1, respectively) were greater than the mean fine OC/EC ratios (3.0:1 and 4.0:1, respectively). The mean coarse OC/EC ratios exceeded mean fine OC/EC ratios during all seasons at JST and YRK, and during summer and autumn at CTR. The high coarse OC relative to EC could hypothetically be related to the presence of vegetation detritus, spores, bacteria, or other noncombustion biological material in the coarse fraction.

Long-term downward trends in $PM_{2.5}$ mass and species concentrations are found at all SEARCH sites (Blanchard et al., 2013). The 1999–2010 mean concentrations were lower than 1999–2001 averages (Hansen et al., 2003) by 0.9–2.7 μg m^{-3} for mass, 0.5–0.9 μg m^{-3} for sulfate (SO_4), 0.2–0.5 μg m^{-3} for ammonium (NH_4), 0.6–1.6 μg m^{-3} for OM, 0.1–0.5 μg m^{-3} for EC, and 0.01–0.2 μg m^{-3} for NO_3. Mean 1999–2010 MMO concentrations were 0.02 μg m^{-3} lower to 0.05 μg m^{-3} higher than the 1999–2001 averages.

Seasonal variations

Table 2 lists long-term seasonal averages, showing the occurrence of seasonal variations in concentrations of constituents associated with primary emissions and also of secondary products of atmospheric chemical reactions. The middle years (2004–2007) of the monitoring record are used for illustration of seasonal variations, because all sites reported measurements of all species during that period. The inland–coastal SO_4 difference was greatest during summer (1.5 μg m^{-3}) and least in winter (0.1 μg m^{-3}) (Table 2). The mean inland and coastal SO_4 concentrations and their seasonal variations are comparable to those reported in Hand et al. (2012) for southeastern sites during the period 2005–2008. The mean summer (June–August) SO_4 concentrations ranged from 4.2 to 6.9 μg m^{-3} (2.8–4.7 μg m^{-3} as SO_2), compared with mean summer SO_2 concentrations of 2.8–14.4 μg m^{-3} (Table 2). SO_2 constituted 48–61% of SO_x ($SO_2 + SO_4$ as SO_2) at nonurban sites and 58–76% of SO_x at urban sites during summer. During winter, SO_2 constituted 78–88% of SO_x at nonurban sites and 80–91% of SO_x at urban sites. Qualitatively comparable summer–winter seasonality has been reported by the U.S. EPA for the CASTNet (U.S. EPA, 2010). More specifically, during 2004–2007 at the four CASTNet sites located within SEARCH states (Figure 1), SO_2 constituted 23–45% of SO_x in summer and 56–81% of SO_x in winter. Although the seasonal pattern is comparable for SEARCH and CASTNet, SO_2 is a lower fraction of SO_x during both seasons at CASTNet sites than at the nonurban SEARCH sites. The nonurban SEARCH mean SO_2 concentrations are higher than mean SO_2 concentrations at the nearest CASTNet sites, possibly due to closer proximity of SEARCH sites to major point sources (Blanchard et al., 2013). Conversion of SO_2 to SO_4 was ~10–80% complete within the SEARCH domain, and depended on season and location.

Table 2. Seasonal mean concentrations, 2004–2007: SO_2 and SO_4 as SO_2; NH_3 and NH_4 as NH_3; NO, NO_2, HNO_3, NO_3, NO_y species as NO_2. Units are $\mu g\ m^{-3}$. The first four sites are coastal. For each site pair, the nonurban site is listed first

Site	Season[a]	SO_2	SO_4	NH_3	NH_4	NO	NO_2	NO_y	HNO_3	NO_3	EC	OM
OAK	Spring	3.5	2.3	0.4	1.0	0.2	2.6	5.2	0.7	0.2	0.5	3.5
	Summer	2.8	3.0	0.2	1.1	0.2	2.1	4.1	0.4	0.1	0.4	4.0
	Autumn	4.4	2.4	0.2	1.0	0.2	3.0	5.4	0.5	0.2	0.5	3.7
	Winter	5.2	1.5	0.2	0.8	0.4	5.3	8.2	0.7	0.4	0.8	4.7
GFP	Spring	3.0	2.3	0.7	1.1	2.3	11.1	16.1	0.2	0.3	0.6	2.9
	Summer	3.9	2.8	0.6	1.1	1.7	12.3	16.7	0.6	0.2	0.6	3.4
	Autumn	6.1	2.5	0.6	1.2	4.4	15.8	23.7	0.5	0.3	0.8	3.7
	Winter	5.9	1.4	0.5	0.8	8.7	16.3	29.1	0.3	0.5	0.7	3.1
OLF	Spring	4.2	2.3	0.5	1.1	1.0	8.1	11.3	0.5	0.3	0.6	3.3
	Summer	4.5	2.9	0.4	1.2	2.3	8.8	13.0	0.5	0.2	0.7	4.1
	Autumn	7.9	2.5	0.3	1.1	1.5	11.3	14.9	0.4	0.2	0.8	4.1
	Winter	7.0	1.4	0.3	0.7	1.8	11.8	15.5	0.2	0.4	0.6	3.1
PNS	Spring	8.4	2.3	0.8	1.2	3.7	14.4	21.1	0.8	0.4	0.6	3.3
	Summer	6.8	3.0	0.6	1.1	3.5	14.4	21.6	0.4	0.2	0.6	3.5
	Autumn	10.9	2.4	0.6	1.1	7.3	18.4	28.8	0.5	0.3	0.7	3.5
	Winter	11.4	1.5	0.6	0.8	11.8	19.7	33.7	0.1	0.4	0.8	3.8
CTR	Spring	4.2	2.3	0.3	1.1	0.3	3.3	6.3	0.9	0.2	0.6	4.9
	Summer	3.5	3.6	0.2	1.4	0.2	2.6	5.1	0.5	0.1	0.5	4.8
	Autumn	6.5	2.7	0.2	1.2	0.6	5.8	8.9	0.4	0.2	0.6	4.1
	Winter	6.9	1.5	0.3	0.8	0.8	7.5	11.1	0.6	0.4	0.6	3.5
BHM	Spring	13.8	2.8	2.4	1.5	23.4	39.5	65.1	0.9	0.6	2.1	7.3
	Summer	14.4	4.6	2.4	2.1	16.5	34.4	55.3	1.0	0.3	1.7	6.9
	Autumn	15.4	2.8	2.1	1.4	32.9	39.2	73.1	0.5	0.4	1.8	5.8
	Winter	13.3	1.7	1.4	1.1	40.2	39.8	81.2	0.4	1.0	1.7	5.4
YRK	Spring	7.4	2.7	1.7	1.5	0.5	6.0	10.8	1.5	0.5	0.6	5.0
	Summer	7.0	4.6	2.5	2.3	0.4	5.7	9.6	1.1	0.3	0.6	5.7
	Autumn	8.1	3.0	2.6	1.6	0.9	9.2	13.7	1.1	0.5	0.6	4.4
	Winter	10.9	1.5	2.0	1.1	1.6	13.0	17.7	1.0	1.0	0.6	3.5
JST	Spring	11.9	2.7	1.3	1.5	34.9	43.3	80.8	1.2	0.5	1.2	6.3
	Summer	10.9	4.7	1.2	2.2	17.0	40.1	61.5	1.2	0.3	1.4	6.1
	Autumn	12.1	3.0	1.2	1.6	49.3	44.2	96.6	0.7	0.5	1.5	5.7
	Winter	15.8	1.6	1.0	1.2	60.3	47.9	107.7	0.5	1.1	1.4	6.2

Notes: [a]Winter = Dec.–Feb.; Spring = March–May; Summer = June–Aug.; Autumn = Sept.–Nov.

Mean $PM_{2.5}$ NO_3 concentrations were highest during winter, but were still only 0.5–0.8 $\mu g\ m^{-3}$ at CTR and the coastal sites, and 1.3–1.5 $\mu g\ m^{-3}$ at BHM, YRK, and JST (Table 2, as NO_3). The 2004–2007 mean $PM_{2.5}$ NO_3 concentrations constituted 1–5% of NO_y in winter and 0.6–3% of NO_y in summer (Table 2).

Mean NO and NO_2 accounted for 54–100% of mean NO_y concentrations, depending on site and season (Table 2), indicating incomplete conversion of NO_x ($NO + NO_2$) to reaction products. Summer 2004–2007 NO concentrations averaged 0.2–0.4 $\mu g\ m^{-3}$ (as NO_2) at rural sites, 1.7–3.5 $\mu g\ m^{-3}$ at Gulf Coast sites, and 17 $\mu g\ m^{-3}$ at BHM and JST (30% of NO_y at BHM and 28% of NO_y at JST). Winter NO concentrations averaged 0.4–1.6 $\mu g\ m^{-3}$ (as NO_2) at rural sites, 2–12 $\mu g\ m^{-3}$ at Gulf Coast sites, and 40–60 $\mu g\ m^{-3}$ at BHM and JST (50% of NO_y at BHM and 56% at JST). NO_2 constituted 62% of NO_y at BHM and 66% of NO_y at JST in summer; in winter, NO_2 constituted 49% of NO_y at BHM and 45% at JST. These seasonal differences in mean NO and NO_2 concentrations at BHM and JST indicate that NO was converted to NO_2 more rapidly in summer than in winter, consistent with the reaction of NO with higher summer concentrations of O_3 to form NO_2.

The 2004–2007 reduced nitrogen (NH_x = sum of NH_3 and NH_4, as NH_3) mean concentrations ranged from 1.0 to 3.1 $\mu g\ m^{-3}$ during winter and from 1.3 to 4.8 $\mu g\ m^{-3}$ during summer (Table 2). NH_4 concentrations were higher in summer than in winter, following competing seasonal variations of SO_4 (1.3–3.1 $\mu g\ m^{-3}$ higher in summer than in winter) and NO_3 (0.2–0.8 $\mu g\ m^{-3}$ higher in winter than in summer) (Table 2, SO_4 as equivalent SO_2 mass and NO_3 as equivalent NO_2 mass). Consequently, NH_3 constituted 23–65% of NH_x during winter and 14–53% of NH_x during summer. Mean NH_3 concentrations were higher at urban than nonurban sites for each site-pair, except JST and YRK (Table 2).

The EC seasonal maximum occurred in the same season as the OM maximum at six of the eight sites (YRK and JST were exceptions), but the sites exhibited seasonal maxima in different seasons (Table 2). The ratio of mean OC/mean TC was modestly lower in winter (0.70–0.82) than in summer (0.74–0.87).

Urban excess concentrations

Mean urban excess concentrations are computed as the differences between the mean concentrations at paired urban and nonurban SEARCH sites: BHM–CTR, JST–YRK, GFP–OAK, and PNS–OLF. Urban excess $PM_{2.5}$ mass concentrations were 2.6–5.0 $\mu g\ m^{-3}$ at inland site pairs but only 0–1.6 $\mu g\ m^{-3}$ at coastal site pairs. $PM_{10\text{-}2.5}$ concentrations displayed an urban excess of 2–9.5 $\mu g\ m^{-3}$ (Table 1).

Mean $PM_{2.5}$ SO_4 concentrations were 3.2–3.4 $\mu g\ m^{-3}$ at coastal SEARCH sites and 3.5–4.3 $\mu g\ m^{-3}$ at inland SEARCH sites; the highest mean SO_4 concentrations were 4.3 $\mu g\ m^{-3}$ at the two urban, inland sites (Table 1). The urban $PM_{2.5}$ SO_4 excess was 0.2–0.8 $\mu g\ m^{-3}$ at inland site pairs and 0.02–0.2 $\mu g\ m^{-3}$ at coastal site pairs (Table 1). Mean NH_x ($NH_3 + NH_4$, as NH_3) concentrations were in the range of 1–5 $\mu g\ m^{-3}$ at SEARCH sites (Tables 1 and 2). Mean NH_x in the cities was larger than at nonurban sites except for YRK, which is influenced by emissions from local poultry production.

Carbonaceous components (EC + OM) represented 47–48% of the average $PM_{2.5}$ mass at BHM and JST, and 36–42% at the other locations (Table 1), approximately the same fraction as $SO_4 + NH_4$ (36–45%). Urban excess carbon (C) concentrations varied geographically and by season, whereas the mean nonurban C concentrations were less variable across the domain (Figure 2). Mean nonurban EC concentrations were 0.5–0.6 $\mu g\ m^{-3}$ at all sites (Table 1), with seasonal variations of 0–0.3 $\mu g\ m^{-3}$ (Table 2). The urban coastal sites exhibited mean EC enhancements of 0.1–0.2 $\mu g\ m^{-3}$ relative to nonurban coastal sites (Table 1), with seasonal variations of 0.2 $\mu g\ m^{-3}$ in their mean concentrations (Figure 2). The urban inland sites exhibited larger EC enhancements of 0.8–1.2 $\mu g\ m^{-3}$ relative to nonurban inland sites (Table 1). Seasonal mean urban excess TC was 2.4–3.2 $\mu g\ m^{-3}$ at BHM, 1.0–2.8 $\mu g\ m^{-3}$ at JST, and –0.5 to +0.6 $\mu g\ m^{-3}$ at PNS (Table 2, Figure 2). OLF is a suburban, rather than rural site, so the concentration difference for PNS–OLF may be negative at times due to variations in local influences around PNS and OLF.

For the sites and time periods having C isotope measurements, modern and fossil components were calculated based on ^{14}C and ^{12}C concentrations as previously described. The mean fossil TC concentrations were 0.5–3.9 $\mu g\ m^{-3}$ and mean modern TC

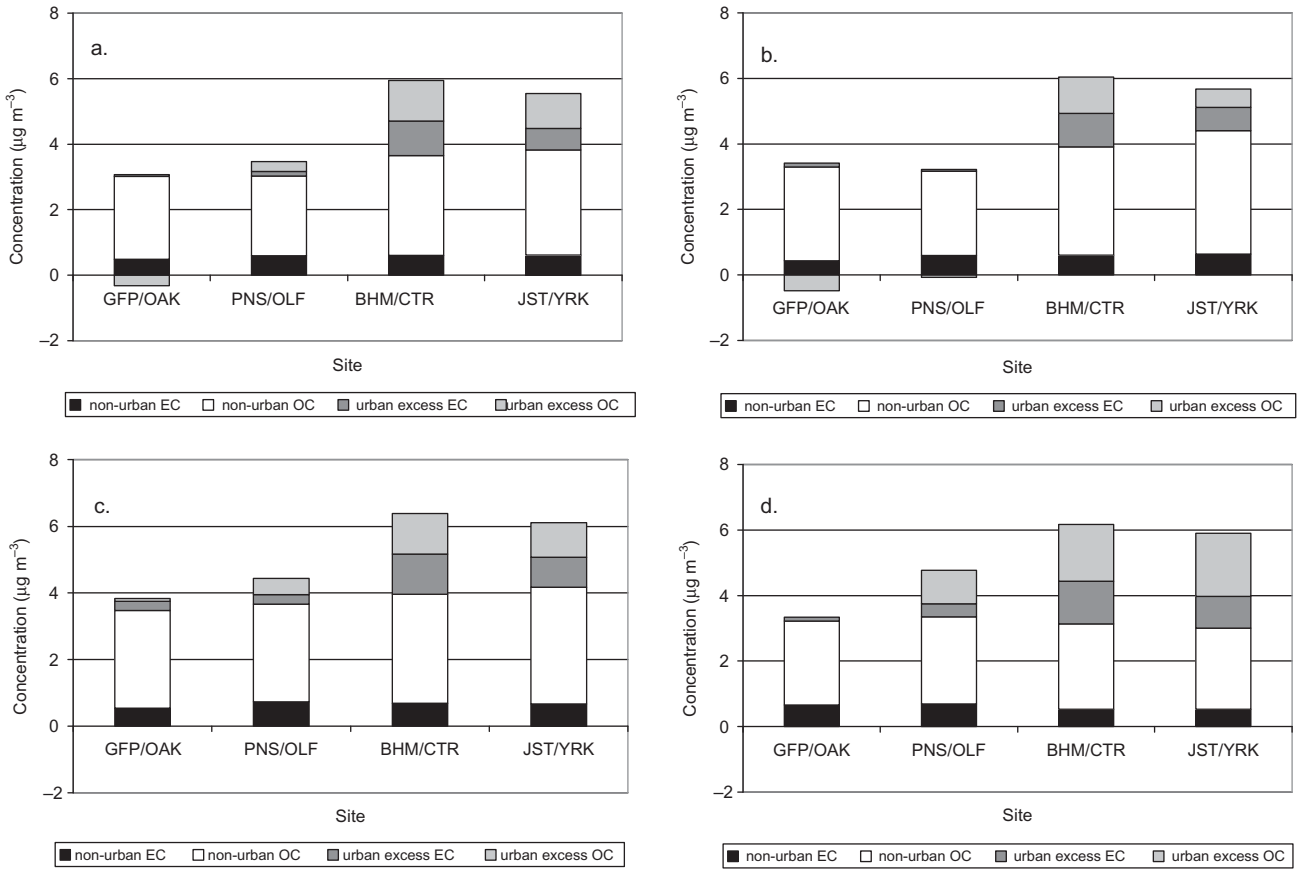

Figure 2. Mean urban excess OC and EC by site pair and season (a) spring (March–May), (b) summer (June–August), (c) autumn (September–December), (d) winter (December–February). The nonurban EC and OC are the mean concentrations at nonurban sites, 1999–2010. Urban excess was computed as the difference between the paired urban and nonurban site means. For spring and summer, mean OAK OC exceeded mean GFP OC.

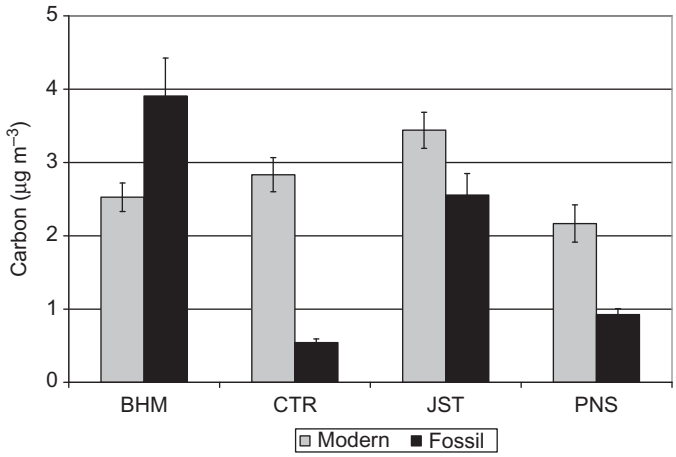

Figure 3. Mean modern and fossil TC. Error bars are one standard error of the mean.

concentrations were 2.2–3.4 μg m^{-3} (Figure 3). At inland sites, the urban excess fossil TC concentrations were 2.0 μg m^{-3} (JST–CTR) and 3.4 μg m^{-3} (BHM–CTR). In comparison, the urban excess modern TC concentrations were smaller: 0.6 μg m^{-3} (JST–CTR) and –0.3 μg m^{-3} (BHM–CTR).

Fine MMO was 3.0 and 5.4% of $PM_{2.5}$ mass at JST and BHM, respectively, but coarse MMO was 37.6 and 34.1% of coarse mass at JST and BHM, respectively. Both fine and coarse mean MMO concentrations were lowest at the nonurban inland sites and highest at the urban inland sites. These differences imply urban MMO contributions of ~ 0.2–0.6 μg m^{-3} for fine PM and ~2–4 μg m^{-3} for coarse PM at BHM and JST (BHM has a larger urban contribution than JST). $PM_{2.5}$ MMO originates from point source and diesel engine emissions, as well as from soil and road dust (Reff et al., 2009; Blanchard et al., 2012). The mean coarse MMO concentrations were ~3–6 times greater than the mean fine MMO concentrations (Table 1), consistent with size distributions expected from dust sources.

Intraurban variations and contrasts

With the exception of O_3, the data in Tables 1 and 2 indicate substantial differences in gas concentrations among SEARCH sites, and relatively smaller differences in particle species concentrations. These intraregion contrasts can be compared with intraurban variations. The two examples of interest where in-city spatial resolution can be obtained by using measurements from different programs are Birmingham and Atlanta.

The BHM site co-hosts instrumentation from the SEARCH, IMPROVE, CSN, and state/EPA compliance networks, providing information on inter-network comparability. Linear regression was used with and without intercept terms to determine the comparability of measurements between networks and between measurement methods. For $PM_{2.5}$ mass and 11 of 19 $PM_{2.5}$ speciation comparisons, systematic measurement differences were less than 10% as indicated by no-intercept regression slopes of 0.9 to 1.1 and r^2 values ranging from 0.85 to 0.98. MMO, EC, and OC exhibited differences related to measurement methods. The SEARCH and IMPROVE networks use the same protocol for partitioning total carbon between EC and OC; CSN switched to the IMPROVE/SEARCH protocol over the period 2007–2009 (U.S. EPA, 2012b). The earlier CSN protocol yields different ratios of OC-to-EC than the IMPROVE protocol (Chow et al., 2001; Conny et al., 2003).

BHM and CTR exhibited the highest and lowest mean $PM_{2.5}$ mass concentrations, respectively, among monitoring sites located in Jefferson County and in the counties surrounding Birmingham (Table 3). Historically, Birmingham has been a major industrial center for producing steel and other metal products. Present industry includes major metal processing industries, with transportation concentrated in the valley of the city. Concentrations of $PM_{2.5}$ mass and some $PM_{2.5}$ species at BHM are likely higher than at other sites in Jefferson County due to the influence of industrial activities situated within 10 km of the BHM monitor (Hansen et al., 2003; see also "Network Description"). CTR, SIPS, and PROV are the most rural monitoring sites near Birmingham, and exhibit the lowest mean concentrations of $PM_{2.5}$ mass and $PM_{2.5}$ species (Table 3).

JST shows urban excess concentrations relative to YRK; however, YRK is at times affected by Atlanta pollution even though it is nominally a rural site. Previous work has shown that local variations among Atlanta monitoring sites are attributable to proximity to highways and variations in wind directions, but JST is representative of air quality in the metropolitan area (Butler et al., 2003). The JST, SDK, or TUC sites exhibit the highest $PM_{2.5}$ mass or species concentrations among Atlanta area sites (Butler et al., 2003) (Table 4). YRK and YG are the most rural PM monitoring sites and exhibit lower mean concentrations of $PM_{2.5}$ mass than the more urban sites (Table 4). Atlanta is larger than Birmingham in area and population, with less topographical variation. Commercial operations and light industry, with a major residential population, characterize Atlanta. Emission sources are largely dominated by transportation, with some stationary sources in the area surrounding the city.

For the majority of gases and $PM_{2.5}$ species, the ranges of mean concentrations occurring within the Atlanta and Birmingham areas were as large as, or larger than, the ranges occurring across the SEARCH network (Tables 1, 3, and 4). In contrast to $PM_{2.5}$ mass and other gases, the highest mean peak 8-hr O_3 concentrations within the Birmingham and Atlanta areas were recorded at the CTR and YRK sites, respectively, located outside the metropolitan areas (Tables 3 and 4). The ratio of mean $PM_{2.5}$/mean PM_{10} ranged from 0.50 (at BHM) to 0.70 (at SIPS and YRK); intermediate ratios of 0.60–0.66 characterized CTR and the urban sites of JST, WYL, and LEED (Tables 3 and 4).

Coefficients of variation (CV) are used here to describe the intersite variability of species concentrations within the Birmingham and Atlanta areas. CVs ranged from 0.02 to 0.15 for species either secondary in nature or influenced by secondary processes (i.e., peak 8-hr O_3, SO_4, and $PM_{2.5}$ mass). CVs were 0.4–1.1 for primary species (CO, NO_y, EC, MMO, and SO_2). The Birmingham CVs for directly emitted species were nearly a factor of 2 greater than Atlanta CVs: NO_y (1.1 vs. 0.6), EC (0.7 vs. 0.3), SO_2 (0.4 vs. 0.2), and MMO (0.5 vs. 0.3) (Tables 3 and 4). This difference is due to the differing topographic conditions of the cities, variations in dispersion, and the concentration of heavy industry in downtown Birmingham, leading to more variability in Birmingham due to source proximity.

Table 3. Mean air pollutant concentrations determined from SEARCH,[a] CSN,[b] IMPROVE,[c] and compliance[d] monitoring sites in Jefferson County, Alabama, and adjacent counties, 1999–2008. The averaging period was selected to provide comparability of records among data sources. Site locations are shown in Figure 1. O_3 is the mean of daily peak 8-hr O_3 from March through October of each year. Other means were compiled from all measurements. Coefficient of variation (CV) = (std. dev.)/mean

	Particles ($\mu g\ m^{-3}$)								Gases (ppbv)			
Site	PM_{25}	SO_4	NO_3	NH_4	EC	OC	MMO	PM_{10}	O_3	SO_2	CO	NO_y
BEAR[d]									50.7			
BES1[d]								27.7				
BESW[d]								25.4				
BHM[abcd]	17.5	4.6	0.9	1.7	1.9	4.7	1.0	35.0	45.7	4.6	420.0	33.2
BM11[d]								27.9				
CORN[d]	14.7								48.4			
CTR[a]	12.0	3.8	0.3	1.2	0.6	3.1	0.4	18.3	50.9	1.8	166.6	4.2
DUNC[d]									44.3			
FAIR[d]								27.3	44.4	4.6	509.9	
FINL[d]											818.8	
HOOV[d]	14.9								46.7			
JASP[d]	14.4											
LEED[d]	15.4							25.9	45.9			
MCAD[d]	15.4								46.4			
PINS[d]	14.9								47.3			
PROV[bd]	13.9	4.3	0.5	1.1	0.5	3.0	0.5		47.6			
SHUT[d]								49.6			996.3	
SIPS[c]	11.6	4.2	0.7	1.3	0.5	2.2	0.6	16.5				
SWEE[d]								36.7				
TARR[d]								28.1	45.9			
WYLA[bd]	17.6	4.5	0.8	1.4	0.9	3.6	1.2	27.6				
Mean	14.8	4.3	0.6	1.3	0.9	3.3	0.7	28.8	47.0	3.7	582.3	18.7
Std. Dev.	1.9	0.3	0.2	0.2	0.6	0.9	0.3	8.6	2.1	1.6	328.5	20.5
CV	0.13	0.07	0.38	0.17	0.67	0.28	0.46	0.30	0.05	0.44	0.56	1.10
N sites	11	5	5	5	5	5	5	12	12	3	5	2
BHM rank[e]	1	1	1	1	1	1	2	3	10	1	4	1
CTR rank[e]	10	5	5	3	3	3	5	11	1	3	5	2

Notes: [a]SEARCH. Values for BHM are from SEARCH. [b]CSN. Co-located with SEARCH at BHM; data used for intercomparison. PROV closed in 2005. [c]IMPROVE. Co-located with SEARCH at BHM; data used for intercomparison. SIPS NH_4 measured 9/2002–2/2005. [d]Compliance monitors. [e]Rank from highest to lowest mean concentration.

Regional variations and meteorological influences

Spatial covariance analysis (principal component analysis, PCA) provides insight into regional geographical variations. In principle, PCA provides a means of identifying independent contributors to the variance of concentration patterns over a spatial domain. In the SEARCH domain, contributors to variance include the nature and location of source emissions, topography, and long-term average meteorological patterns. Applied to single pollutants, the method is also known as empirical orthogonal functions (EOF), and has previously been used to delineate subregions within a network of monitoring sites (e.g., Hidy, 1994). We use PCA here as an objective means for differentiating among sites that may exhibit different influences from weather and from the distribution and nature of source emissions. PCA was applied individually to concentrations of $PM_{2.5}$ mass, $PM_{2.5}$ species, and gas-phase species, since each pollutant may exhibit unique source-receptor characteristics or meteorological dependences. For each species, each principal component (PC) identifies a group of sites having daily concentrations that co-vary over the period of SEARCH operations (Table 5). The results group the sites geographically, by type (e.g., coastal vs. inland or urban vs. rural), or in more complex combinations, depending on the species analyzed. The PCs for $PM_{2.5}$ mass, peak 8-hr O_3, SO_4, NH_4, NO_3, and MMO concentrations differentiated the network into coastal and inland sites (Table 5). The coastal–inland division identified by PCA indicates different inland influences on $PM_{2.5}$ mass concentration, MMO, and secondary species (peak O_3, SO_4, NH_4, NO_3) compared with the Gulf Coast, but does not identify whether meteorological factors or locations of sources cause the differences. CTR appears to be a transitional location, as it was associated slightly more with coastal sites for MMO, slightly more with inland sites for SO_4 and mass, and split for peak O_3.

The spatial groupings for OM and EC also exhibited geographical variation, but were more complex than for the species described immediately above. The four inland sites appear as a

Table 4. Mean air pollutant concentrations for monitoring sites in Fulton County (Atlanta), Georgia, and surrounding counties, 1999–2008. The averaging period was selected to provide comparability of records among data sources. Site locations are shown in Figure 1. O_3 is the mean of daily peak 8-hr O_3 from March through October of each year. Other means were compiled from all measurements. Coefficient of variation (CV) = (std. dev.)/mean

	Particles ($\mu g\ m^{-3}$)								Gases (ppbv)			
Site	PM_{25}	SO_4	NO_3	NH_4	EC	OC	MMO	PM_{10}	O_3	SO_2	CO	NO_y
CNF[d]									51.6	3.0		
CNY[df]									51.1			
COW[e]									43.9			
FT[g]	14.0	4.1	1.0	1.6	0.9	4.5						
GAS[e]									51.4			
GAT[d]	14.9									3.4		
JST[a]	16.3	4.6	0.9	1.8	1.4	4.4	0.5	24.7	51.8	4.6	452.5	45.2
SDK[bcdf]	16.5	4.5	0.7	1.4	1.1	5.3	0.5		49.3		545.3	44.6
SDK[g]	14.4	3.9	0.8	1.5	1.1	4.8						
SND[e]									52.6			
TUC[dfg]	15.4	4.4	1.1	1.8	0.8	4.9			50.2			26.8
YG[g]	12.9	3.8	0.9	1.5	0.5	4.5						
YRK2[f]	15.2								52.1		288.5	
YRK[a]	13.5	4.4	0.8	1.8	0.6	3.1	0.3	19.2	54.6	2.9	182.3	6.6
Mean	14.8	4.2	0.9	1.6	0.9	4.5	0.4	22.0	51.4	3.5	367.1	30.8
Std. Dev.	1.2	0.3	0.1	0.2	0.3	0.7	0.1	3.9	2.8	0.8	162.7	18.3
CV	0.08	0.07	0.15	0.10	0.34	0.15	0.27	0.18	0.06	0.23	0.44	0.59
N sites	9	7	7	7	7	7	3	2	11	4	4	4
JST rank[h]	2	1	3	1	1	6	1	1	5	1	2	1
YRK rank[h]	8	4	5	1	6	7	3	2	1	4	4	4

Notes: [a]SEARCH. PM_{10} measurements are 1999–2007. [b]CSN. SDK is mean of CSN and IMPROVE data. [c]IMPROVE. SDK is mean of CSN and IMPROVE data. [d]Compliance monitors. [e]CASTNet. [f]PAMS. [g]ASACA (Trail, 2010). TUC closed in 2007, mean of 1999–2006 observations. YG commenced in 2002, mean of 2002–2008 observations. [h]Rank from highest to lowest mean concentration.

group for OM, as they were for SO_4 and $PM_{2.5}$ mass. Three (urban and suburban) of the four coastal sites are grouped for OM, but the fourth (rural) site (OAK) associates uniquely with a different (third) PC. For EC, three coastal sites (OLF, PNS, GFP) are associated with PC1; three inland sites (BHM, JST, YRK) are associated with PC2, OAK is uniquely associated with PC3, and CTR is uniquely associated with PC4 (Table 5). The PC3 and PC4 groupings appear to be associated with the occurrence of biomass combustion events whose influence at both OAK and CTR has been observed directly by interpreting time series of PM and gas-phase measurements (Edgerton et al., 2001, 2004). Unlike the simpler coastal–inland division for secondary species (peak 8-hr O_3, SO_4, NH_4, NO_3), the more complex geographic differentiation of EC and OM could be caused by variations in a combination of combustion sources and biogenic influence as well as persistent differences between coastal and inland weather.

Three site groupings were obtained for NO_y (Table 5). These groups represent differences in source influence, as indicated by differences in mean NO_y concentrations (lowest at rural sites, highest at inland urban sites; Table 1). The four PC site groups for SO_2 are distributed geographically (Table 5): PC1 is common to three of the four inland sites (JST is the exception), PC2 and PC4 are associated with the coastal sites, and PC3 is at JST. This pattern suggests local influences for SO_2 that are distinct from the predominant influences for SO_4 and NO_y.

The relationships of the PCA site groups to meteorological influences are examined next. Since PCs are linear combinations of the measurements, a PC score was computed for each date as the daily site concentrations multiplied by the values listed in Table 5; for example,

$$\begin{aligned} PC2_{mass} = {} & 0.27 \times OAK_{PM^*} + 0.23 \times GFP_{PM^*} + 0.32 \times OLF_{PM^*} \\ & + 0.25 \times PNS_{PM^*} + 0.75 \times CTR_{PM^*} + 0.82 \times BHM_{PM^*} \\ & + 0.92 \times YRK_{PM^*} + 0.91 \times JST_{PM^*} \end{aligned} \tag{1}$$

The variables (OAK_{PM^*}, etc.) in eq 1 are standardized values of the daily $PM_{2.5}$ mass concentrations at each site, that is, (daily $PM_{2.5}$ – site mean)/(site standard deviation). As indicated in eq 1, the value obtained for $PC2_{mass}$ on any given date is high when CTR_{PM^*}, BHM_{PM^*}, YRK_{PM^*}, and JST_{PM^*} are high. Similarly, high PC2 scores for peak O_3, SO_4, NH_4, and NO_3 occur when concentrations of those species are high at the inland sites. In contrast, PC1 scores for $PM_{2.5}$ mass, peak O_3, SO_4, NH_4, NO_3, OM, EC, and MMO are highest when concentrations of those species are highest at the coastal sites (Table 5).

PC2 scores were computed for each species and date, and these data were averaged by deciles. As shown in Figure 4, the inland PC (PC2) scores for mass, OM, peak O_3, and SO_4 each increased with

Table 5. Principal component (PC) analysis of $PM_{2.5}$ mass, $PM_{2.5}$ species, and gas-phase concentrations using all available measurements, 1999–2010. Analyses are based on 24-hr resolution data (except for peak daily 8-hr O_3) and utilize days having data from all sites. Tabled values are the correlation of each site concentration with each PC. PCAs were conducted separately for each species and the PCs are species dependent. Italicized values identify the PC having the highest correlations with each site's species concentrations

			Coastal sites				Inland sites			
Species	PC	Var%[a]	OAK	GFP	OLF	PNS	CTR	BHM	YRK	JST
Mass	PC1	66	*0.77*	*0.90*	*0.89*	*0.88*	0.50	0.38	0.20	0.19
	PC2	17	0.27	0.23	0.32	0.25	*0.75*	*0.82*	*0.92*	*0.91*
SO_4	PC1	76	*0.84*	*0.89*	*0.90*	*0.88*	0.55	0.40	0.30	0.29
	PC2	13	0.40	0.31	0.35	0.34	*0.74*	*0.84*	*0.92*	*0.89*
MMO	PC1	67	*0.89*	*0.91*	*0.91*	*0.87*	*0.66*	0.25	0.33	0.20
	PC2	15	0.31	0.22	0.26	0.33	0.59	*0.73*	*0.84*	*0.89*
NH_4	PC1	73	*0.86*	*0.91*	*0.91*	*0.88*	0.58	0.40	0.26	0.24
	PC2	15	0.36	0.27	0.33	0.31	*0.71*	*0.83*	*0.92*	*0.90*
NO_3	PC1	64	*0.74*	*0.87*	*0.84*	*0.84*	0.42	0.32	0.24	0.26
	PC2	14	0.36	0.29	0.21	0.37	*0.72*	*0.82*	*0.85*	*0.86*
OM	PC1	51	0.33	*0.68*	*0.80*	*0.91*	0.16	0.34	–0.01	0.24
	PC2	18	0.14	0.11	0.22	0.20	*0.66*	*0.74*	*0.87*	*0.84*
	PC3	8	*0.84*	0.51	0.35	0.09	0.48	0.20	0.16	–0.09
EC	PC1	48	0.35	*0.77*	*0.73*	*0.85*	0.19	0.44	–0.07	0.41
	PC2	14	0.12	0.19	0.21	0.16	0.30	*0.66*	*0.83*	*0.77*
	PC3	11	*0.86*	0.23	0.17	0.01	0.15	–0.12	0.28	–0.11
	PC4	7	0.15	–0.02	0.33	0.23	*0.90*	0.21	0.19	0.03
Peak O_3	PC1	75	*0.86*	*0.87*	*0.90*	*0.89*	*0.65*	0.36	0.39	0.25
	PC2	13	0.33	0.32	0.34	0.33	*0.64*	*0.87*	*0.85*	*0.92*
CO	PC1	48	*0.65*	*0.84*	0.55	*0.75*	0.27	0.37	–0.07	0.14
	PC2	16	0.55	0.02	*0.60*	0.20	*0.80*	0.09	*0.88*	0.17
	PC3	10	–0.03	0.28	0.20	0.37	0.11	*0.77*	0.16	*0.88*
SO_2	PC1	39	0.32	0.03	0.06	0.14	*0.42*	*0.79*	*0.75*	0.10
	PC2	14	0.08	*0.78*	0.24	*0.85*	0.39	–0.05	0.23	0.13
	PC3	11	0.09	0.02	0.10	0.14	0.20	–0.08	0.22	*0.96*
	PC4	9	*0.78*	0.35	*0.80*	0.04	*0.43*	0.26	0.07	0.15
NO_y	PC1	51	0.03	0.29	*0.66*	0.57	0.11	*0.71*	0.20	*0.85*
	PC2	14	0.52	0.07	0.45	0.09	*0.60*	–0.04	*0.90*	0.17
	PC3	11	*0.68*	*0.85*	0.22	*0.68*	*0.60*	0.49	0.05	0.06

Note: [a]Percent variance explained by unrotated PCs.

increasing temperature and with decreasing transport distance (length of 24-hr back-trajectory computed using HYSPLIT). Therefore, spatial coherence of inland PM and its two largest components (OM and SO_4) and the spatial coherence of peak 8-hr O_3 were driven by meteorological conditions favoring conversion of primary to secondary species (temperature) and limited horizontal advection (stagnation). Volatility of organic compounds may affect the more complex relationship of the inland OM PC to temperature (e.g., mean temperature does not increase from the second highest to the highest decile OM).

The coastal PCs (PC1) for mass, OM, and SO_4 did not exhibit consistent monotonic relationships to temperature or to transport distance. However, PC1 for mass, OM, and SO_4 increased as relative humidity (RH) decreased. In addition, PC1 for OM increased as the difference between the 850 millibar (mb) and the surface temperatures became less negative (from –6 K to 0 K), suggesting that coastal OM increased as vertical mixing became more limited. Segregation of PCs by direction of transport suggested a distinction between easterly to southerly winds relative to winds from the north. The coastal PCs showed strongest positive relations with northerly or northeasterly winds, with declining PC values for winds from the south and north-northwest. The PCs associated with MMO displayed a strong influence in transport distance between 320 and 580 km, but showed little relation with transport direction, suggesting a domain-wide baseline for MMO.

Regionwide occurrences of elevated PM concentrations

Many studies have documented persistent elevated concentrations of $PM_{2.5}$ mass in the Southeast, including within the

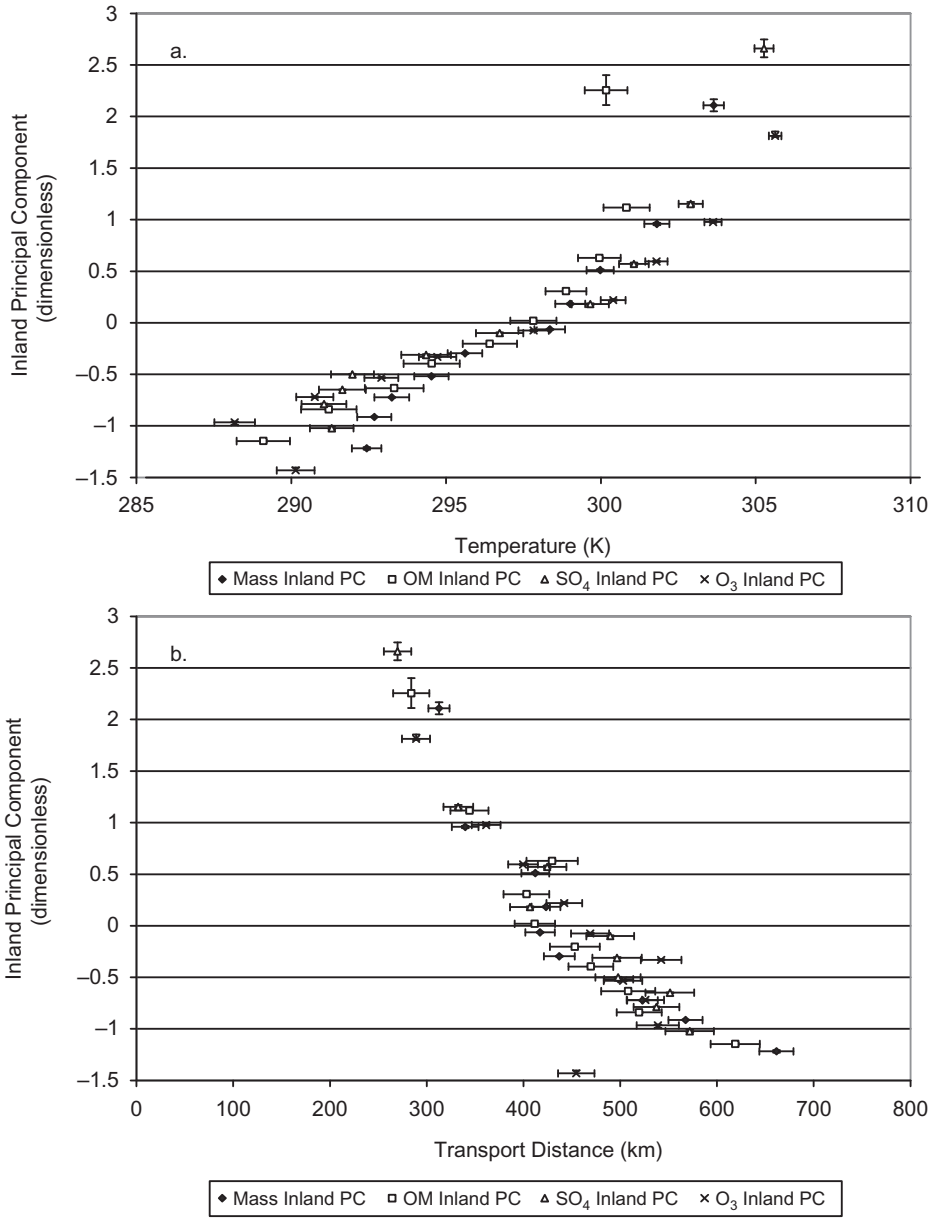

Figure 4. Decile mean inland PC (PC2) versus (a) daily maximum temperature at Birmingham Airport and (b) 24-hr HYSPLIT back-trajectory distance from Birmingham Airport. Error bars are one standard error of the mean and are smaller than the symbols when not visible.

SEARCH domain, and elsewhere in the eastern United States. The regional aerometric conditions in the Southeast that are associated with elevated concentrations of $PM_{2.5}$ mass, and comparison with conditions further north, are thus of interest. To this end, three measures of regionwide occurrence of elevated $PM_{2.5}$ mass concentrations were examined: (1) cases where all eight SEARCH sites exhibited high $PM_{2.5}$ mass or species concentrations, (2) cases where the four inland sites exhibited high $PM_{2.5}$ mass or species concentrations, and (3) cases where only coastal sites exhibited high concentrations.

Table 6 lists the numbers of days when high (95th percentile or greater) species concentrations occurred at SEARCH sites. Since not all species were measured at all sites during all years, the actual number of dates when all eight sites exceeded 95th percentile concentrations is potentially larger than shown; however, the data record suffices to identify example dates for further study. Assuming that the data collected capture most of the regional and local events, regional-scale pollution events (days with values exceeding 95th percentile concentrations) involving more than 6 of the 8 SEARCH sites are rare (<2% of days); subregional events affecting 4–6 sites occur on ~10% of days. On five dates (comprising three episodes), $PM_{2.5}$ mass concentrations exceeded the site 95th percentiles at all eight sites; concentrations exceeding 95th percentiles occurred at all operational sites (6 or 7 sites) on an additional six dates. Network-wide (all operational sites, at least 6 sites operating) high concentrations occurred on 16 dates for MMO, 9 dates for SO_4, 8 dates for NH_4, 7 dates for NO_3, and no dates

Table 6. Number of days when one or more SEARCH sites had concentrations exceeding annual site 95th percentiles, 1999–2010. The 95th percentiles were determined for each species, site, and year. The dates with concentrations exceeding 95th percentile values at any site were identified and the number of sites exceeding 95th percentile values on each date were tabulated. The PM data are 24-hr concentrations obtained by filter-based sampling. A total of 4369 days data had PM mass concentrations from one or more sites. The numbers of days with PM species concentrations ranged from 4199 to 4308. The gas-phase data are continuous hourly measurements compiled as peak daily 8-hr maxima for each species; in total, 4371 days had data from one or more sites

	Number of monitoring sites exceeding site annual 95th percentile									
Species	1	2	3	4	5	6	7	8	All[a]	Total
Mass	340	169	103	60	24	13	10	5	11	719
SO_4	218	83	44	29	13	16	5	6	9	408
NO_3	260	82	37	28	12	10	8	4	7	437
NH_4	247	83	49	21	18	5	6	3	8	429
OM	354	111	39	18	6	6	1		0	535
EC	383	121	33	18	5	3			0	563
MMO	252	74	43	35	18	19	18	5	16	459
O_3	312	186	89	40	16	9	4		0	656
SO_2	759	168	48	6	2	1			0	984
NO_y	598	169	60	14	6	1	1		1	849
CO	646	177	56	10	1				0	890

Note: [a]All operational sites when at least 6 sites were operating (i.e., 6 of 6, 7 of 7, or 8 of 8).

for either EC or OM (Table 6). The dates for SO_4 include a multiday episode in 1999 and two dates falling within multiday high PM episodes.

Network-wide high PM mass or high SO_4 days occurred primarily during the warmest months (June through September), and were characterized by the presence of a high barometric pressure system over the southeastern United States, with clear skies over most or all of the SEARCH domain and precipitation along the Atlantic Coast, north of the Ohio River, or west of the Mississippi River (NOAA, 2010). Synoptic-scale, stagnating anticyclonic meteorological patterns have previously been linked to regional SO_2 and SO_4 events in the midwestern to northeastern United States (Hidy, 1994). In the SEARCH domain, HYSPLIT back-trajectories (Draxler et al., 2009) exhibited mean 24-hr transport distances of ~250 km when $PM_{2.5}$ mass was high (>95th percentile) at 7–8 sites (16 days) compared with mean 24-hr distances of ~500 km when all sites exhibited $PM_{2.5}$ mass concentrations less than 95th percentile values (3610 days) (Figure 5). The results in the SEARCH domain indicate the significance of persistent regional stagnation conditions, most frequently occurring in summer with a stationary or slowly moving frontal system within an eastward-moving high barometric pressure regime. This situation contrasts with conditions favoring long-range transport, as previously described for the northeastern United States (e.g. Hidy, 1994).

Daily meteorological conditions were further examined for days during 1999–2007. In those years, high (95th percentile or greater) $PM_{2.5}$ mass concentrations occurred at all four inland sites without co-occurrence of high $PM_{2.5}$ mass concentrations at coastal sites on 13 dates (there were 47 dates with 3 or 4 of the 4 inland sites having high PM; 30 of these 47 days had high PM at CTR and 45 of the 47 days had high PM at YRK). Each of the 13 inland-wide PM events occurred during the months of May through October; 8 of the 13 dates were in the months of June through August. All 13 inland-wide events were associated with high barometric pressure (979–994 mb) at Atlanta Hartsfield Airport (1014–1031 equivalent sea-level pressure). Transport distances calculated from HYSPLIT 24-hr back trajectories were less than 100 km on 6 dates and less than 300 km on 9 of the 13 dates. Most of the 13 days were characterized by the presence of a high barometric pressure system over the southeastern United States, and precipitation along the Gulf Coast (NOAA, 2010). On one date (August 15, 2007), high pressure was centered off the coastline near North Carolina and a tropical depression was located offshore over the Gulf of Mexico; on another date (September 11, 2003), an inland high-pressure system extended southwest from the state of New York to the

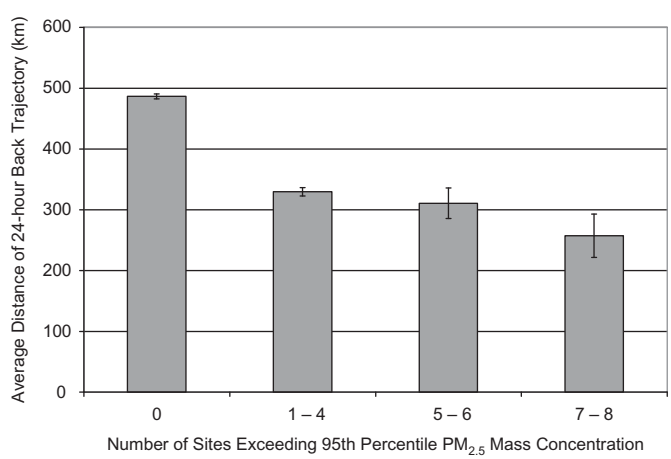

Figure 5. Average 24-hr transport distance determined from HYSPLIT back trajectories versus number of SEARCH sites having daily $PM_{2.5}$ mass concentration values exceeding site-specific 95th percentile values. Error bars are one standard error of the mean.

Gulf of Mexico and a hurricane ("Ophelia") was located over the western Atlantic Ocean. The inland high-PM events therefore exhibit stagnation conditions affecting inland areas, but not necessarily the Gulf Coast.

High (95th percentile or greater) $PM_{2.5}$ mass concentrations occurred at one or more coastal sites without co-occurrence of 95th percentile or greater $PM_{2.5}$ mass concentrations at inland sites on 212 dates from 1999 to 2007. On 36 days of this series, high $PM_{2.5}$ mass concentrations occurred at 3 or 4 of the 4 coastal sites. These days were characterized by the presence of a low barometric pressure system over or close to the SEARCH domain (including tropical depressions off the Atlantic coast on August 3, 2004, and over the SEARCH domain on September 15, 2002), and a stationary or approaching cold front. These conditions evidently follow from southward penetration of low-pressure systems from the north and west. Such systems frequently terminate synoptic scale stagnation associated with high-pressure systems. Superimposed on pervasive mesoscale sea–land breezes along the Gulf of Mexico, these conditions allow pollutant accumulation ahead of the approaching front.

Discussion

SEARCH measurements and complementary data from other monitoring programs expand current understanding of the chemical climatology of aerosols in the southeastern U.S. and improve the characterization of urban/nonurban and inland/coastal contrasts. This work complements large-scale modeling studies and analyses of data from national monitoring programs (e.g., Phillips and Finkelstein, 2006). The distinction between regionally characteristic pollutants and those affected by local conditions is an important aspect of air-pollutant exposure in the Southeast. Covariation of O_3 and $PM_{2.5}$ mass and composition across multiple SEARCH sites is driven by synoptic-scale meteorological influences, especially elevated temperature and air mass stagnation. Spatial variations across the SEARCH network are partly due to differences between inland and coastal conditions previously indicated in modeling studies and in analyses of IMPROVE data (e.g., Malm et al., 2002, 2004; Hand et al., 2012), and partly due to source-related differences between urban and nonurban settings. Urban excess concentrations and intraurban variations are attributable to differences in emission source proximity and areal density of emissions as characterized in other studies such as the U.S. EPA supersite program for the St. Louis, MO, region (Turner and Allen, 2008) and the New York City area (Demerjian and Mohnen, 2008).

The importance of local source emissions is reinforced by observed local urban enhancements of primary emitted species, CO and NO_y. The mean nonurban CO concentrations reflect a regional baseline level of ~100–200 ppbv, increasing by factors of ~2–4 in the cities. The urban NO_y enhancement is as large as an order of magnitude. These enrichments affirm that the local urban atmospheric environment differs in important ways from regional conditions. Smaller urban enhancements of SO_2 are evident for the urban/nonurban site pairs, especially BHM–CTR, indicating the effects of local SO_2 emission sources superimposed on a regional baseline driven by nonurban sources of SO_2 nominally located within the region.

In contrast to the urban enhancements of CO, NO_y, and SO_2, the mean O_3 concentrations were 2.3–13.2 ppbv higher at nonurban sites than at the paired urban locations. Mean peak 8-hr O_3 concentrations were lower at BHM and JST (24.7 and 24.5 ppbv, respectively) than at other locations (29.1–37.7 ppbv). Urban NO emissions react with ambient O_3 to decrease peak O_3 concentrations at BHM and JST and form NO_2; summer NO_2 concentrations are over 60% of mean NO_y at BHM and JST, but winter NO_2 concentrations are 45–50% of NO_y.

The urban excess in coarse mass, $PM_{10\text{-}2.5}$, is especially pronounced at BHM, indicating the importance of local emission sources. The urban excess $PM_{2.5}$ mass concentrations provide opportunities for emission reductions from air quality management strategies. However, the regional baseline of 11–12 $\mu g\ m^{-3}$ is high relative to the new federal annual $PM_{2.5}$ national ambient air quality standard (NAAQS) of 12 $\mu g\ m^{-3}$. This presents a challenge to further reducing fine particle concentrations in population centers of the Southeast.

The mean urban excess SO_4, NH_4, and NO_3 concentrations are 0.02–0.8 $\mu g\ m^{-3}$, 0.05–0.4 $\mu g\ m^{-3}$, and 0.1–0.5 $\mu g\ m^{-3}$, respectively, and vary seasonally due to the influence of meteorological processes and chemistry. The urban excess concentrations are greatest in summer for NH_4 and in winter for NO_3, consistent with expectations based on temperature-dependent thermodynamic equilibrium between SO_4, NH_4, HNO_3, and NO_3 (Blanchard and Hidy, 2003, 2005; Blanchard et al., 2007).

The average carbonaceous component, representing 36–48% of the mean $PM_{2.5}$ mass, could be larger if the average OM/OC exceeds the value of 1.4 adopted as a convention by the SEARCH network. Modern carbon is an important component of $PM_{2.5}$ mass at all locations and likely is both anthropogenic (e.g., biomass combustion) and biogenic (including primary OC from fires or entrainment of detritus, and secondary OC derived from atmospheric oxidation of biogenic emissions of VOCs). Mean modern TC concentrations are more uniform (2.2–3.4 $\mu g\ m^{-3}$) than mean TC (3.2–6.9 $\mu g\ m^{-3}$), suggesting the importance of widespread area sources of modern TC of biological origin and more localized point or mobile sources of fossil TC.

Since primary EC emissions are accompanied by primary OC emissions in equal or greater amounts for the majority of emission source types and in total (Chow et al., 2010), the BHM and JST mean urban excess EC (1.2 and 0.8 $\mu g\ m^{-3}$, respectively) suggests the existence of urban, primary OC contributions averaging over 1 $\mu g\ m^{-3}$ (potentially representing primary urban OM of 1.4-2 or greater at BHM and JST). Primary urban EC and OM at BHM and JST is potentially reducible by targeting emissions of primary carbonaceous material from urban sources.

Mean MMO concentrations are a small fraction of $PM_{2.5}$ mass. MMO concentrations are higher in summer at all sites, especially at coastal locations. The summer mean MMO coastal enhancement reverses the coastal–inland gradient that occurs in the mean annual concentrations of other measured species. The reason for this difference is not known, but an MMO gradient along the coast could be related to the presence of pervasive low levels of African Sahara dust in summer. Alternatively, gradients

could be associated with agricultural operations or unidentified sources within the SEARCH domain. The MMO gradient does not decrease from west to east, as might be expected of dust arriving from arid areas of Texas or Mexico.

Network-wide days of high PM mass or SO_4 concentrations occurred primarily during the warmest months (June through September). Enhanced $PM_{2.5}$ mass, OM, O_3, and SO_4 concentrations at inland sites are favored by increased atmospheric oxidant concentrations and reduced wet deposition, associated with increasing temperature, decreasing transport distance, and stagnation. The presence of secondary OM is complicated by a trade-off between the reactivity of particle producing VOCs, increasing with temperature and oxidant concentration, versus increased volatility of VOCs and their oxidation products with increasing temperature. This complexity is embedded in the seasonal variability of OM concentrations, and has been identified previously for secondary organic aerosol formation at JST (Blanchard et al., 2011).

Summer synoptic conditions most often involve the presence of large-scale anticyclonic, high-barometric-pressure systems over the southeastern United States, clear skies over most or all of the SEARCH domain, and precipitation along the Atlantic Coast, north of the Ohio River, or west of the Mississippi River. Persistent stagnation often lasting more than a day with nocturnal inversion capping promotes cumulative increases in concentrations of all pollutants over the Southeast until wind speeds increase along the frontal system, convection enhances dilution through vertical mixing, and precipitation decreases pollution concentrations. These air mass stagnation conditions are distinct from the combined stagnation and air mass transport sequence that occurs further north with summer frontal systems moving from west to east (Hidy, 1994). The occurrence of regionwide, elevated pollution levels in the SEARCH domain involving more than six SEARCH sites is rare (<2% of days). High (95th percentile or greater) $PM_{2.5}$ mass concentrations occurred at coastal sites independent of high $PM_{2.5}$ mass concentrations at inland sites on days characterized by the presence of a low barometric pressure system over or close to the SEARCH domain, and a stationary or approaching cold front penetrating southward to the Gulf Coast.

Conclusions

SEARCH measurements and complementary data from other monitoring programs expand current understanding of the chemical climatology of aerosols in the southeastern United States. The measurements provide insight into local pollution levels embedded within the regional domain. Regional distribution of sources and unique meteorological influences lead to high average pollution levels. Regional conditions affect urban pollution inland, but less so along the Gulf Coast. In general, the long-term results are consistent with and reaffirm earlier SEARCH data analyses based on shorter time records. The regional influence is greatest with secondary species, including O_3, particulate SO_4, and particulate organic matter, some of which is produced by atmospheric oxidation of VOCs from vegetation and anthropogenic sources. Other species, many of which are from primary sources, are more influenced by local sources. Of the various pollutants of interest measured to date in the Southeast, atmospheric carbon in the gas and condensed phases remains least well characterized climatologically, in terms of both sources and the differentiation of the secondary material produced in the atmosphere. Limited measurements of modern and fossil total carbon point to the importance of biological and biogenic emissions in the Southeast in making up a sizeable fraction of airborne particles.

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