ATLANTA AUTOMOTIVE PARTICULATE MATTER EXPOSURE AND EVALUATION

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ATLANTA AUTOMOTIVE PARTICULATE MATTER EXPOSURE AND EVALUATION

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NOMENCLATURE

ACE: Atlanta Commuter's Exposure Study
Ag: Silver
Al: Aluminum
ASACA: Assessment of Spatial Aerosol Composition in Atlanta
As: Arsenic
B: Boron
Ba: Barium
BC: Black Carbon
Ca: Calcium
Cd: Cadmium
Co: Cobalt
CPC: Condensation Particle Counter
Cu: Copper
EC: Elemental Carbon
EF: Enrichment Factor
Fe: Iron
FID: Flame Ionization Detector
GPS: Global Positioning System
HRV: Heart Rate Variability
IC: Inorganic Carbon
K: Potassium
Li: Lithium
Mg: Magnesium
Mn: Manganese
Mo: Molybdenum
Na: Sodium
Ni: Nickel
OC: Organic Carbon
OPC: Optical Particle Counter
P: Phosphorus
Pb: Lead
Pd: Palladium
PM: Particulate Matter
$PM_{2.5}$: Particulate Matter with a diameter less than 2.5 μm
PM_{10} : Particulate Matter with a diameter less than 10 μm
PUF: Polyurethane Foam
Rb: Rubidium
S: Sulfur
Sb: Antimony
Se: Selenium
Sn: Tin
SOA: Secondary Organic Aerosol

TD-GCMS: Thermal Desorption-Gas Chromatography/Mass Spectrometry
TEOM: Tapered Element Oscillating Microbalances
Ti: Titanium
TOC: Total Organic Carbon
TOT: Thermal Optical Transmittance
U: Uranium
V: Vanadium
WSOC: Water Soluble Organic Carbon
Zn: Zinc

SUMMARY

The following thesis titled, Atlanta Automotive Particulate Matter Exposure and Evaluation, presents data obtained as a part of a joint project with Emory University, Rollin's School of Public Health. The Atlanta Commuters Exposure (ACE) Study uses both real-time and time-integrated sampling techniques for ambient aerosol concentrations. The ACE study is unique in that it will correlate the ambient aerosol concentrations with the concurrent health measurements. The primary objective of this thesis is to measure the concentration, size distribution and the chemical composition of $PM_{2.5}$ inside the vehicle cabin for several commuters. The vehicles followed a scripted route along roadways in the Atlanta metropolitan region during periods of peak traffic volume, while the compact air sampling package of both real-time and time-integrated instruments recorded data. Real-time measurements for Particulate Matter (PM) were made using compact Optical Particle Counters (OPC), a Condensation Particle Counter, and a MicroAethalometer. The time-integrated measurements for Elemental Carbon (EC), Organic Carbon (OC), Water Soluble Organic Carbon (WSOC), particulate elemental concentrations, and speciated organics required filter collection methods. Thus a compact air-sampling package was created to combine both sets of real-time and time-integrated instruments. The following results are presented for the first four commutes. The framework for analyzing and presenting results is developed, and will be used for future commutes.

CHAPTER 1: INTRODUCTION

Mobile source emissions based on fossil fuel combustion have been the focus of many recent research studies (Brunekreef, 2007; Carrico, 2003; Gehring, 2002; Hoek, 2002; Lanki, 2006; Lough, 2005; Peters, 2005; Peters, 2004; Schauer, 1996; Singh, 2004; Weber, 2007). One of the most important environmental impacts of these emissions seen in the recent century relates to ambient air quality and its impact on human health (Brunekreef, 2002; Curtis, 2006; Dockery, 1996; Gauderman, 2004; Gehring, 2002; Gold, 2005; Hoek, 2002; Lanki, 2006; Oftedal, 2008; Peters, 2006; Peters, 2005; Peters 2004; Pope, 1995). Ice core data has clearly demonstrated a link between anthropogenic emissions and poor air quality in the Northern Hemisphere (McConnell, 2007; Simoes, 2001). Understanding air quality impacts on human health is important for devising emission standards and guidelines. Thus, analysis of air pollutants is vital for both from a human health and global ecosystem perspective.

Ambient aerosol classification is an important step in understanding and accounting for various lung and cardiovascular health problems (Brunekreef, 1997; Gauderman, 2004; Gehring, 2002; Lanki, 2006). Studies have shown that ambient fine particulate matter ($PM_{2.5}$, particles with an aerodynamic diameter less than or equal to 2.5 micrometers) concentrations above a certain value are associated with increased adverse health effects (Brunekreef, 2001; Curtis, 2006; Dockery, 1993; Gold, 2005; Pope, 1995). Specifically, it has been found that children who live in urban settings have a significantly greater risk of developing asthma (Brunekreef, 1997; Gauderman, 2004; Gehring, 2002; Oftedal, 2008). The traffic networks of

Interstate 75, Interstate 85, State Route 400, and Interstate 285 are dispersed throughout the metro Atlanta area and are a prime location to study $PM_{2.5}$ levels and the associated adverse health effects of fossil fuel combustion. To understand specifically how the urban setting and interstate networks are affecting population exposures to particulate matter, we need to quantify the amounts, chemical composition, and sources of $PM_{2.5}$.

$PM_{2.5}$ has many sources in the Atlanta area including biogenic, power plant, and mobile source emissions (Odman, 2009; Weber, 2003; Zheng, 2007). Previous studies have shown a significant relationship between fine particulate matter and cardiovascular health abnormalities (Peters, 2004; Peters, 2005). However, specific sources of the PM, which contribute to these health problems, have not been fully characterized. Thus, there is a stronger need for using source appointment to estimate the contributions of sources to ambient PM levels.

Recently there has been a greater correlation with traffic-related $PM_{2.5}$ emissions and adverse health conditions, including cardiovascular problems, heart rate variability, and myocardial infarctions (Dockery, 1993; Peters, 2004; Peters, 2005; Pope, 1995). Exposure studies in other countries included people exposed to the traffic either from operating mass transit (i.e. bus drivers, taxi drivers, etc.) or from living in proximity to major roadways (Hoek, 2002; Lanki, 2006; Peters, 2004). Another study has found that there are elevated heart rates in elderly people when they are riding on mass transit lines, while being exposed to high $PM_{2.5}$ values from the surrounding traffic (Gold, 2005). Along with these specific studies, there have been laboratory animal exposure studies in a controlled environment that show the

aforementioned adverse health problems (Singh, 2004). One potential problem with animal studies results from the introduction of far greater $PM_{2.5}$ levels than are typically seen in urban environments, and specifically close to roadways. The ultimate issue arises from attempting to quantify $PM_{2.5}$ to which people are exposed with real-time and time-integrated measurements. The complex measurements for Elemental Carbon (EC), Organic Carbon (OC), Water Soluble Organic Carbon (WSOC), elemental concentrations, and speciated organics cannot be made using a compact real-time air-sampling package.

In this thesis, data are presented that were obtained as a part of a joint project with Emory University, Rollin's School of Public Health. The Atlanta Commuters Exposure (ACE) Study uses both real-time and time-integrated sampling techniques for ambient aerosol concentrations. The ACE study is unique in that it will correlate the ambient aerosol concentrations with the concurrent health measurements.

CHAPTER 2: ATLANTA COMMUTERS EXPOSURE STUDY

The Atlanta Commuters Exposure (ACE) Study, a joint effort by both Emory University, Rollin's School of Public Health, and the Georgia Institute of Technology, attempts to deal with the problems of real-time data acquisition of $PM_{2.5,}$ speciation, and concurrent health effects. The primary objective of the Atlanta Commuters Exposure study is to "investigate the exposure of automobile-commuters in the Atlanta region to hazardous air pollutants and to assess changes in indicators of cardiopulmonary status such as heart rate, heart rate variability (HRV), and biomarkers of airway and systemic inflammation and oxidative stress." The primary objective of this thesis is to measure the concentration, size distribution and the chemical composition of $PM_{2.5}$ inside the vehicle cabin for several commuters. The vehicles followed a scripted route along roadways in the Atlanta metropolitan region during periods of peak traffic volume, while the compact air sampling package of both real-time and time-integrated instruments recorded data. The goal is to do this while not altering the in car environment enough to change the health or comfort of the driver.

The test subjects in this study are limited to 20 healthy adults, and 20 adults who have been diagnosed by a physician as having Asthma. The preliminary test subjects have been volunteers, and work for the Georgia Institute of Technology, Emory University, or the Center for Disease Control. Future test subjects will be selected from a pool previously involved in a transportation engineering study (the Commute Atlanta Study) by Dr. Randall Guensler of Georgia Institute of

Technology. A list of exclusion criteria for test subjects is enumerated in Appendix A. In this thesis preliminary data is presented for the first four commutes. The framework for analyzing and presenting results is developed, and will be used for future commutes.

CHAPTER 3: INSTRUMENTATION

3.1 Instrumentation Overview

In-cabin particulate measurements are divided into two main categories, time-integrated and real-time instruments. These two sets of measurements allow understanding of the overall exposure of subjects, and the time/location of extreme exposures. This will give insight into ambient situations that can cause the largest dose of pollutants to the test subjects. Descriptions of the two different instrument sets follows. A schematic for the instrument measurement system is shown below in Figure 3.1 and an overview of the measurements being made are detailed in Table 3.1.

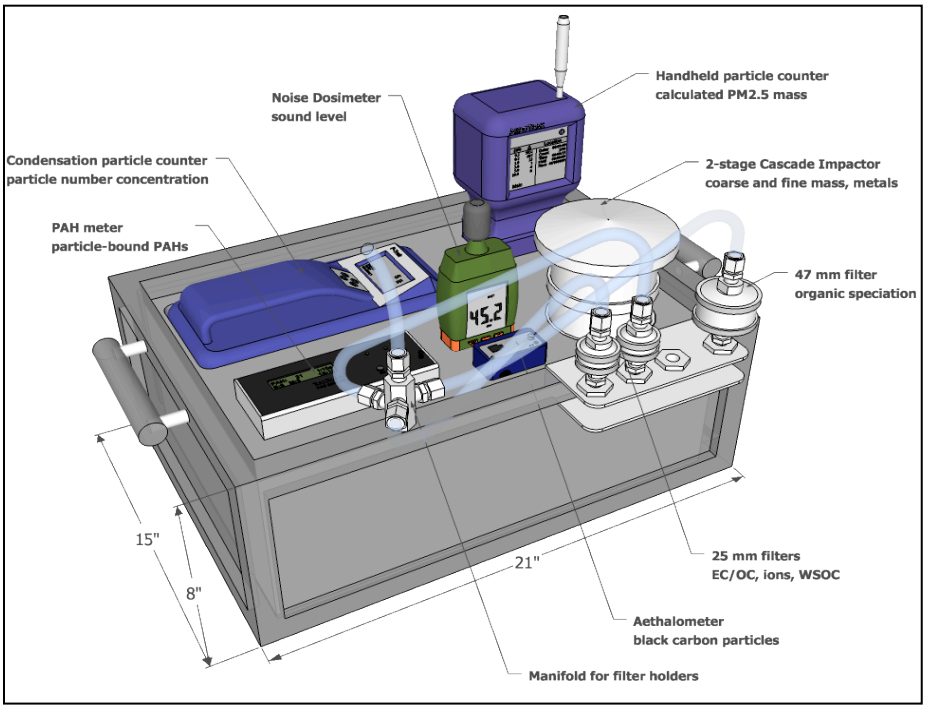

Figure 3.1: Portable sampling package with real-time and time-integrated instruments (Roby Greenwald, Emory University)

Table 3.1: Measurements and associated sampling technique

		Detection Method	Measurement Time Scale	Power Source
Integrated measurements (filter-based)				
Coarse and fine mode mass	co-sampled	Gravimetric	Time-integrated	external
Trace elements ($PM_{2.5}$)	co-sampled	ICP-MS	Time-integrated	external
Organic speciation		TD-GC/MS	Time-integrated	external
EC/OC ($PM_{2.5}$)	co-sampled	TOT	Time-integrated	external
WSOC ($PM_{2.5}$)	co-sampled	TOC	Time-integrated	external
Ionic species ($PM_{2.5}$)	co-sampled	IC	Time-integrated	external
Real-time measurements				
$PM_{2.5}$ volume concentration		OPC	1 Second	internal
Light-absorbing aerosols ($PM_{2.5}$)		Filter transmittance	1 Minute	internal
Number Concentration (Ultra Fine PM)		CPC	1 Second	internal

3.2 Time-integrated Measurements

3.2.1 Filter Based Carbon Speciation

Time-integrated samplings for Elemental Carbon (EC), Organic Carbon (OC), and Water Soluble Organic Carbon (WSOC) were completed with two 25 mm pre-baked Quartz filters run side-by-side. Both filters were held in place with a Pall Corporation Stainless Steel Filter holder that have inline $PM_{2.5}$ cut point impactors. All filter handling was carried out at the Georgia Institute of Technology inside a clean room with the associated clean room attire. Preceding each commute, to obtain blank values, the filter holders were loaded with pre-baked filters identical to those being used for sampling. The filters were immediately removed to take filter-handling contamination into consideration. The sample filters were then loaded into the filter holders and then transported in sterile bags to Emory University to be

installed in the test subject's car. Flow rates as well as times that the pumps were connected and disconnected were recorded for the filters. Investigations determined that a flow rate of 15 L/min through each filter allowed maximum air sampling capability with the available pump strength. After sampling, the filter holders and filters were returned to Georgia Institute of Technology and unloaded in a clean room into baked foil lined Petri dishes.

Analysis of the filters for EC, OC, Water Soluble Organic Carbon (WSOC), and ion concentrations began by splitting the two filters in half with a 25 mm filter half-punch tool. Half of the filters were used for EC/OC and the other half were used for WSOC and Ionic concentrations collectively. The methodology behind halving two different filters was the need to remove variability due to the short sampling time and low flow rate. Tests were run on the Georgia Institute of Technology's campus both on the roof of the Ford Environmental Science and Technology building and next to Interstate 75/85 in downtown Atlanta to validate this approach.

After the two 25 mm filters were split into two sections, the EC/OC measurements were made using a Sunset Laboratory Thermal Optical Transmittance (TOT) analyzer. This instrument used a 1 cm^2 filter punch size with the methodology of Thermal Optical Transmittance (TOT). This method heats the filter in a helium environment completely absent of oxygen so that the organics on the filter volatize. The organics are then oxidized to form CO_2 and then sent through a methanator oven, which changes the CO_2 to methane. The methane is then measured with a Flame Ionization Detector (FID) and treated as the Organic Carbon. Next, oxygen is added to the pure helium environment and the elemental carbon on the filter is oxidized to

$CO_{2,}$, which is then converted to methane and measured by the FID to determine the elemental carbon content. During this entire process, the pyrolitic conversion is monitored by the transmittance of a laser through the filter sample and the values are compared to determine EC and OC values. The reported EC and OC concentrations, which were then blank adjusted, were converted to corresponding OC/EC ratios were calculated for further analysis.

The WSOC samples were analyzed starting with an aqueous extract method to get all water-soluble compounds into the aqueous phase. The half- samples were placed into two separate glass jars that were cleaned per ACE protocol with 30 mL of ultra-pure DI water. The jars where then sonicated for one hour and placed in refrigeration at 40 degrees Fahrenheit until the WSOC instrument was used for analysis. The WSOC analysis was quantified using the Sievers 900 Portable TOC Analyzer with an in-line quartz fiber filter to make sure only water-soluble compounds were analyzed. WSOC standard analysis were routinely performed and a standard calibration curve is given in Appendix B.

After calibration and aqueous extraction, the WSOC values were measured in ppb, and then converted based on pump flow-rates and sampling time. Since the low relative flow rates of the two 25 mm filters, when compared to the 47 mm filters, the two 25 mm values were averaged to remove any variability. To allow for greater analysis a WSOC/OC ratio was determined.

The remaining aqueous extract solution was then used for Ion Chromatography (IC) analysis; however, results have yet to be converted to ambient concentrations and will be analyzed in the future.

3.2.2 Filter Based Organic Speciation

Organic Speciation of $PM_{2.5}$ was carried out with a 47 mm Quartz filter. The filter was held in place with a Pall Corporation Stainless Steel Filter holder. The handling of the filter holder, as well as transportation, and blank value generation of the filter followed the same procedures as the two 25 mm filter holders previously discussed. A flow rate of 30 L/min was determined to be an adequate sampling volume for the analysis to be carried out on the filter. After sampling was completed the filter was placed in a pre-baked foil lined Petri dish and then stored in a -10 C freezer. Analyses were conducted at the University of Wisconsin-Madison in the laboratory of Dr. James Schauer. The analysis carried out for Organic Speciation utilized the Thermal Desorption-Gas Chromatography/Mass Spectrometry (TD-GC/MS) technique. This includes a thermal desorption by heating the filter in a controlled state and allowing the organic species to volatize off the filter surface. The organics are then separated using gas chromatography and then a mass spectrometer is used to identify a wide range of organic compounds.

3.2.3 Filter Based Elemental Speciation

The elemental time-integrated data methodology included a two-stage cascade impactor for fine and coarse particulate element concentrations. A Harvard two-stage

impactor was used to separate the PM_{10} and $PM_{2.5}$ respectively. The first stage has a jet that makes coarse particles impact onto a Polyurethane Foam (PUF) filter. The $PM_{2.5}$ can however go around the PUF and get deposited onto a pre-cleaned Teflon filter. The Teflon filters were cleaned by the University of Wisconsin-Madison using a nitric acid wash and transported in sealed Petri dishes with Teflon tape. A flow rate of 30 L/min is needed for the two-stage impactor in order to keep the critical flow high enough to separate the coarse and fine PM. Handling and blank values were generated using the same technique as previously discussed for the 25 mm and 47 mm quartz filters. After sampling, the Teflon filters were repackaged in Petri dishes and sealed with Teflon tape. The samples were then put into -10 C freezer before being sent to the University of Wisconsin-Madison for analysis.

Analysis of the Puff and Teflon filter for elements, carried out by the University of Wisconsin-Madison, included using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) described in Lough et. al (2000). This method of ICP-MS incorporates a reduction reaction in a plasma field to reduce the metals on the filter to basic atom form followed by mass spectrometry to identify the atoms. After elemental concentrations were recorded, detection limits were determined for each element. Any values, which were below the detection limit, were not used for further analysis.

3.3 Real-time Measurements

Real-time data acquisition has been divided between three different instruments. The TSI AeroTrak ($PM_{2.5}$), TSI P-Trak (Ultra Fine Particulate Matter),

and a MicroAethalometer (Black Carbon) will be used to measure different aspects of the ambient PM. Each instrument will allow for further understanding of exposure time-scales as well as being able to associate geographic location with maximum exposure concentrations. A list of time resolution of each instrument can be seen in the previously mentioned Table 3.1.

3.3.1 Optical Particle Counter

The TSI AeroTrak is a compact Optical Particle Counter (OPC), which collects data based on the light scattering by particles. The size distribution concentrations are estimated for specific particle size bins. The measured bins from the AeroTrak OPC outputs a number distribution, which can be converted to a volume concentration as follows:

$$V_c = (\pi/6)*D_p^3*N_c$$

(Where D_p is the average optical diameter of the AeroTrak Sampling bins, N_c is the number concentration, and V_c is the volume concentration)

The AeroTrak data was converted from a number concentration to a volume concentration using only the lowest three bins of the AeroTrak (0.3-0.5μm, 0.5-0.7μm, 0.7-1μm). Using the smallest three bins removes the uncertainty associated with the larger bins and the associated size distribution. This method was verified through previous studies of compact OPC size distributions. The values of the AeroTrak volume concentrations were then changed to 5-minute averaged data over the roughly 2-hour commutes.

3.3.2 Condensation Particle Counter

The TSI P-Trak is a portable Condensation Particle Counter, which is used to measure Ultra Fine Particulate Matter with a particle diameter ranging from .02 μm to 1 μm. This instrument works by having an internal cut point of less than 1 μm and then using an alcohol vapor the ultra fine particles are "grown" so that they can be detected and quantified more accurately by an internal OPC. The resulting number concentration is then output by the P-Trak in one-second time integrals and later converted to 5 minute averaged data. This data was not however analyzed with the real-time data later discussed in the thesis.

3.3.3 Aethalometer

For real-time black carbon measurements the Magee Scientific MicroAeth AE51 compact Aethalometer was used. The AE51 collects data using a proprietary Teflon-coated borosilicate glass fiber filter, replaced for each commute, and the Aethalometer optical absorption method. This method quantifies black carbon based on transmittance of light through a sample (Bond, 1999). An internal 880 nm wavelength light source measures the sample on the filter relative to a clean area on the filter. The absorption coefficient is calculated based on the transmittance, and then based on flow rate and sample filter area the BC concentration is recorded in mg BC/m^3 (Quincey, 2009). The concentrations were then changed to 5-minute averaged data to correspond with the other real time instruments.

CHAPTER 4: RESULTS

4.1 Foreword

This section describes the first four of the ACE study commutes for the time-integrated measurements and three of the first ACE study commutes for the real-time instruments. The first commutes, given the ID of HE01, HE03, HE05, and HE07, were conducted during December of 2009. The following results were used for initial estimates of exposure values and methodology evaluation. These values were then compared to typical metro Atlanta values to see the comparison of the values to the typical urban measurements.

4.2 Filter Based Time-integrated Values

4.2.1 Filter Based Carbon Speciation

The EC and OC speciation was carried out the University of Wisconsin-Madison. The EC and OC values for 4 test subjects are located in Table 4.1. The values show that the OC is the dominant form of carbon on the filters during the ACE commutes. This is expected in a typical urban setting (Lim, 2003; Carrico, 2003). In this study we have seen the values range from 27.4 $\mu g/m^3$ to 46.6 $\mu g/m^3$ for OC while previous studies saw typical average OC values for the summer months of 7.7 $\mu g/m^3$ (Carrico, 2003) and 10.82 $\mu g/m^3$ (Weber, 2003). These values are much higher than typical winter ambient OC values. OC generally has higher values in the summer according to yearly data from the Assessment of Spatial Aerosol Composition in Atlanta (ASACA) study (Trail, 2010). The ASACA study has seen

typical urban OC winter values between 2.7μg/m^3 and 4.5 μg/m^3 (Trail, 2010). However, the seasonal variability in OC seems to be very high in rural areas and minimal in urban areas according to the ASACA data. This data leads to the assumption that during the ACE study, the numbers and proximity to mobile sources on the highways has led to the higher OC values which is expected. Also the high OC values, which are attributed to mobile sources, could be the cause for less seasonal variability in OC values in the Atlanta metro area.

Table 4.1: Elemental and Organic Carbon concentrations

Sample ID	OC	OC unc.	EC	EC unc.	OC/EC Ratio
	ug/m3	ug/m3	ug/m3	ug/m3	
ACE-HE01	25.99	1.30	4.12	0.21	6.32
ACE-HE03	46.19	2.31	4.78	0.39	9.65
ACE-HE05	38.42	1.92	2.70	0.22	14.2
ACE-HE07	33.71	1.69	10.61	0.62	3.18
ACE Average	36.08	n/a	5.55	n/a	6.50
Carrico et al. (2003)	7.7	n/a	0.8	n/a	9.6
Weber et al. (2007)	10.82	n/a	3.61	n/a	3.0

The ACE study EC values on average were higher than both the Carrico et al. (2003) and Weber et al. (2007) studies as seen in Table 4.1. These values show a trend of increasing EC with proximity to the highway as the Carrico et al. (2003) study was at the Jefferson Street sampling site and the Weber et al. (2007) study was conducted next to the downtown Atlanta interstate 75/85. The higher number of diesel mobile sources on the I-285 corridor with respect to the other two sampling sites is the reason for the elevated EC values in the ACE study.

OC/EC ratios are also seen in Table 4.1 for each of the four commutes along with the OC/EC ratios of the Carrico et al. (2003) and the Weber et al. (2007) studies. The recorded values in the ACE study shows an average OC/EC ratio of 6.5, which is in between the Weber et al. (2007) value of 3.0 and the Carrico et al. (2003) value of 9.6. The ACE value also falls within the yearly average OC/EC range of 4.5-9.4 for the urban ASACA measurement sites (Trail, 2010). When compared to the Carrico et al. (2003) study, the lower ACE OC/EC ratios are likely due to the sampling time of the ACE study being in the winter rather than the summer as well as the different sampling proximities to mobile diesel sources. Higher OC values seem to dominate the ACE values while the Weber et al. (2007) study OC only reaches 30 percent of the average ACE values even though measurement occurred very close to the interstate. The reason for this is unknown but could be attributed to different traffic concentrations during the two studies.

Table 4.2: WSOC Measurements and WSOC/OC ratios

Sample ID		WSOC Water concentration (ppb)	WSOC Air concentration (ug/m3)	Sample Average WSOC Air Concentration (ug/m3)	WSOC/OC Ratio
ACE-HE01	QU1	82.0	3.03	2.81	0.102
	QU2	67.0	2.58		
ACE-HE03	QU1	167.0	6.14	5.65	0.121
	QU2	136.0	5.15		
ACE-HE05	QU1	314.0	12.3	10.6	0.299
	QU2	212.0	8.88		
ACE-HE07	QU1	198.0	5.90	6.15	0.170
	QU2	186.0	6.40		
ACE Average		170.3	6.30	6.30	0.17
Weber et al. (2007)		n/a	n/a	4.73	0.44

The average ACE OC value, 36.1 μg/m3, is much higher than yearly ambient OC at the urban Fire Station 8 and rural Fort Yargo monitoring sites, 4.57 μg/m3 and 4.49 μg/m3 respectively, according to ASACA (Trail, 2010). The WSOC that is associated with the OC is the next analysis to be reviewed as WSOC can give more insight into Secondary Organic Aerosols (SOA) and the elevated OC seen in the ACE study compared to the ASACA values (Weber, 2007).

The WSOC air concentrations, seen in Table 4.2, were found to be on average higher than the average ambient WSOC values of Weber et al. (2007). However the WSOC/OC ratio for the ACE studies reaches an average value of .17, which is considerably lower than the Weber et al. (2007) study WSOC/OC ratio of .44. This is most likely from the relatively short time allowed for particle aging and thus no time for WSOC to form. This also suggests no correlation between increases in OC and increases in WSOC values. In other recent studies there has been no direct correlation between WSOC and mobile sources and is proposed that WSOC is more of a regional characteristic of the Southeast (Weber, 2007). These data show that there is little time for secondary WSOC to form and most likely does not contribute substantially to of the elevated OC values in the ACE study.

4.2.2 Filter Based Elemental Analysis

The elemental concentrations are located in Table 4.3 and are low when compared to the OC and WSOC values. However, to get a better understanding of the element concentrations relative to those of the surrounding natural environment,

Enrichment Factors (EF) were generated. The EF values are related to the average crustal values for the measured elements.

The enrichment factors were calculated using aluminum as the reference element. Aluminum was chosen because of its relative high crustal concentration and its contribution to $PM_{2.5}$ is assumed to be solely from the Earth's crust (Hagler, 2007). The EF was calculated using $EF= (X_i/Al)_{sample}/(X_i/Al)_{crust}$, where Xi is the selected element of study. The EF values are in Figure 4.1 for the elements Pd, Mg, Rb, Ca, and U all have an EF less than 10. This leads to the assumption that these elements are crustal sources. However, B, Zn, Cd, Cu, As, Sn, Mo, S, Ba, Ag, Sb, and Se which all have EF greater than 1000 can be attributed to anthropogenic sources.

Table 4.3: Filter Based Elemental Concentrations

Sample ID	ACE-HE01	ACE-HE03	ACE-HE05	ACE-HE07
Units	ng/m^3	ng/m^3	ng/m^3	ng/m^3
OC	2.74E+04	4.66E+04	3.55E+04	3.62E+04
WSOC	2.81E+03	5.65E+03	1.06E+04	6.15E+03
Lithium (Li)	1.56E-01	1.69E-01	1.23E-01	1.05E-01
Boron (B)	1.42E+00	1.30E+00	1.73E+00	6.16E+00
Sodium (Na)	7.06E+01	1.02E+02	5.56E+01	3.96E+01
Magnesium (Mg)	9.64E+00	1.42E+01	1.55E+01	8.79E+00
Aluminum (Al)	1.92E+01	2.39E+01	2.10E+01	2.26E+01
Phosphorus (P)	1.07E+01	9.56E+00	7.18E+00	1.12E+01
Sulfur (S)	3.73E+02	4.04E+02	1.50E+02	3.67E+02
Potassium (K)	2.12E+01	3.51E+01	3.24E+01	6.60E+01
Calcium (Ca)	3.52E+01	6.64E+01	4.77E+01	5.94E+01
Titanium (Ti)	5.50E+00	1.96E+01	1.42E+01	8.87E+00
Vanadium (V)	2.51E-01	3.78E-01	7.99E-01	4.02E-01
Manganese (Mn)	1.13E+00	3.37E+00	1.25E+00	2.86E+00
Iron (Fe)	9.62E+01	3.79E+02	9.95E+01	2.49E+02
Cobalt (Co)	3.16E-02	5.30E-02	7.17E-02	4.49E-02
Nickel (Ni)	1.07E-01	6.78E-01	4.57E-01	5.56E-01
Copper (Cu)	9.47E+00	4.30E+01	1.37E+01	1.82E+01
Zinc (Zn)	7.66E+00	1.80E+01	6.54E+00	1.43E+01
Arsenic (As)	3.63E-01	3.34E-01	5.41E-01	1.34E+00
Selenium (Se)	4.30E-01	8.50E-01	8.17E-01	1.38E+00
Rubidium (Rb)	4.26E-02	8.98E-02	7.72E-02	1.61E-01
Molybdenum (Mo)	6.50E-01	1.40E+00	6.03E-01	1.07E+00
Palladium (Pd)	3.13E-03	1.32E-02	6.43E-03	1.22E-02
Silver (Ag)	1.95E-01	6.61E-02	3.34E-02	2.90E-02
Cadmium (Cd)	2.93E-02	4.29E-02	3.76E-02	7.27E-02
Tin (Sn)	7.53E-01	2.85E+00	1.08E+00	9.85E-01
Antimony (Sb)	9.52E-01	4.03E+00	1.47E+00	2.84E+00
Barium (Ba)	4.99E+00	2.90E+01	8.90E+00	1.52E+01
Uranium (U)	3.31E-03	4.08E-03	2.04E-03	3.85E-03
Lead (Pb)	1.20E+00	1.04E+00	9.86E-01	1.71E+00

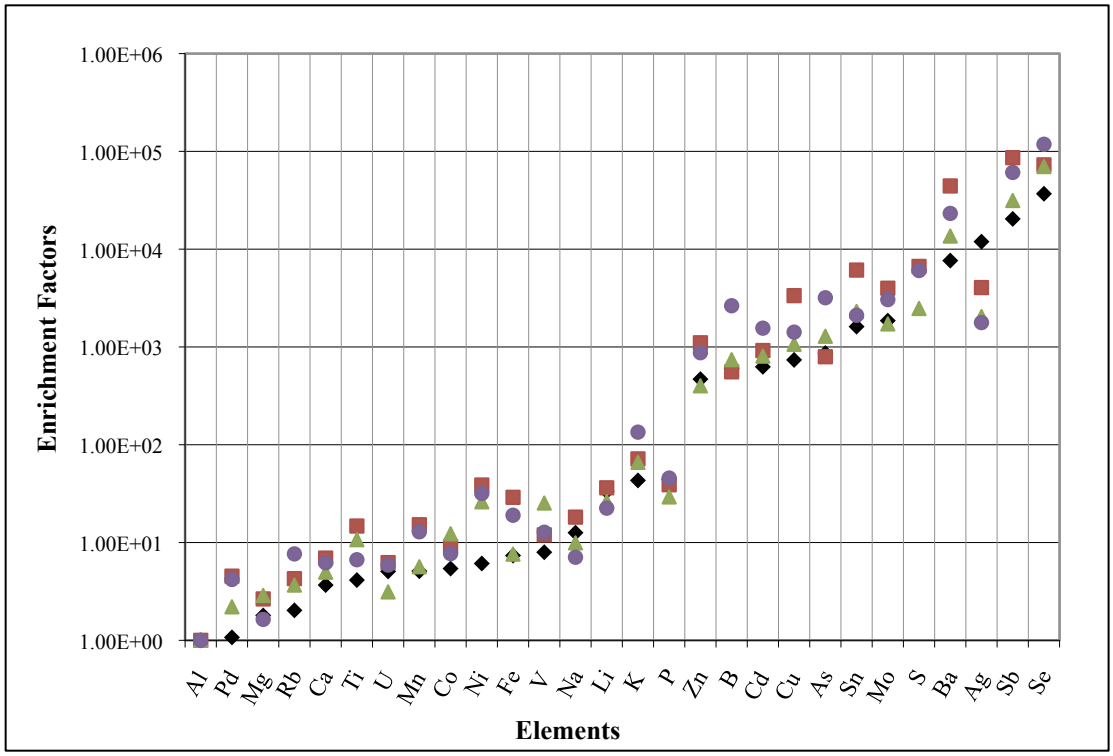

Figure 4.1: Enrichment Factor values of measured elements

The majority of the elements with EF values greater than 1000 can be associated with brake wear, engine emissions, or fossil fuel combustion. Selenium is related to combustion of fossil fuels (Natusch, 1974). Selenium could also be accredited to tire wear as it has been used for rubber vulcanization. Sulfur is largely from the coal power plants around the metro Atlanta area and diesel fleet vehicles. Copper, zinc, barium and antimony can all be associated with brake wear as there were many times during the different commutes where traffic was consistently below the speed limit because of congestion (Lough, 2005). Many of the brake wear elements are typically in the larger particle size range of the $PM_{2.5}$ that were collected and further analysis would be needed to analyze the actual size appointment of the different elements, although it is highly likely brake wear impacts $PM_{2.5}$ elements measured in the ACE study (Lough, 2005).

4.2.3 Filter Based Organic Speciation Analysis

Specific organic compounds measured in the ACE study are presented in Table 4.4. Given the short sampling time the detection of 35 organic compounds supports the methodology of the ACE sampling procedure. Specifically many of the higher concentrations seen in the ACE identified organics can be associated with mobile sources using a study conducted by Schauer et al. (1997) on organic compound appointment. The Schaeur et al. (1997) study can link several ACE identified organic compounds such as tetracosane, tricosane, pentacosane, hexacosane, eiocosane, hopane, and norhopane to diesel or gasoline combustion engines, tire wear, brake wear, and/or paved road dust.

In specific the tetracosane group have relative high values in all of the ACE tests. These compounds can be associated with diesel, catalytic gasoline, and non-catalytic gasoline engine emissions (Schauer, 1996). Elevated values of eicosane were observed specifically during the HE07 test. Eicosane is typically from paved roadway dust, and it is interesting to note that this is the only test where that compound has an elevated value. This reason for this is unknown even though the commutes occurred on the same stretch of highway.

Other organic compounds such as hentriacontane had elevated levels in the ACE tests. This compound is linked to both cigarette smoke and vegetative detritus (Schaeur, 1996). Although these sources are not mobile source related they still represent test subjects' exposures and would be associated to any concurrent health effects.

Table 4.4: Organic Species concentrations (*benzo(b)flouranthen and benzo(k)flouranthen are summed together)

Organic Species	ACE-HE01	ACE-HE03	ACE-HE05	ACE-HE07
	ng/m^3	ng/m^3	ng/m^3	ng/m^3
pyrene -d10				
Phenanthrene	34.40	53.99	32.12	41.96
Anthracene	13.51	123.26	19.27	74.32
Fluoranthene	100.40	811.12	201.34	532.79
Pyrene	50.89	105.60	42.11	59.02
benz(a)anthracene-d12				
Benzo(ghi)fluoranthene	9.65	20.95	11.10	41.48
Cyclopenta(cd)pyrene	14.90	29.16	273.92	848.21
Benzo(a)anthracene	18.34	23.80	10.25	67.04
Chrysene	16.46	31.81	20.85	82.26
Benzo (b)fluoranthene *	52.23	80.34	48.25	372.99
Benzo (k)fluoranthene *				
Benzo(e)pyrene	14.98	36.27	12.57	109.15
Benzo(a)pyrene	22.85	50.09	24.95	293.08
Perylene	14.16	0.00	0.00	82.16
coronene-d12				
Indeno(123-cd)pyrene	15.78	20.39	9.86	139.19
Benzo(ghi)perylene	27.92	34.92	10.68	175.94
Coronene	19.28	6.21	0.60	75.76
20R, aaa - cholestane - d4				
17A(H)-22,29,30-trisnorneohopane	0.00	22.93	21.20	0.00
17A(H)-21B(H)-29-Norhopane	46.79	72.48	27.28	118.90
17a(H)-21b(h)-hopane	28.84	58.67	15.19	69.22
eicosane - d42				
Nonadecane	139.56	0.00	0.00	321.98
Eicosane	164.69	66.63	85.07	4662.40
Heneicosane	298.22	131.13	86.30	247.80
Docosane	660.45	260.21	236.89	531.14
tetracosane-d50				
Tricosane	388.72	877.39	718.23	1121.57
tetracosane	407.15	1614.71	911.21	2493.92
Pentacosane	208.92	1455.20	180.14	970.67
Hexacosane	170.78	1821.30	424.00	317.96
Heptacosane	488.86	871.17	455.64	310.25
triacontane-d62				
Octacosane	335.34	502.29	75.78	158.49
Nonacosane	277.18	291.31	310.19	604.08
Triacontane	323.05	923.04	264.15	464.66
dotriacontane-d66				
Hentriacontane	587.28	1136.28	475.62	1111.89
Dotriacontane	324.20	240.57	243.97	491.13
Tritriacontane	106.02	452.27	346.16	669.35
Tetratriacontane	191.77	216.17	220.82	129.68
Pentriacontane	0.00	379.15	126.08	0.00

When approaching the organic concentrations in graphical form, Figure 4.2, it appears that for the first four tests the higher the molecular weight of the organic the higher the corresponding sample concentration. This shows that the higher molecular weight compounds are more abundant in the sample $PM_{2.5}$. The analysis of more test data should clarify which organic compounds have a consistent higher than normal regional concentrations. This will allow for more stable source appointment based on the Schaeur et al. (1996) study.

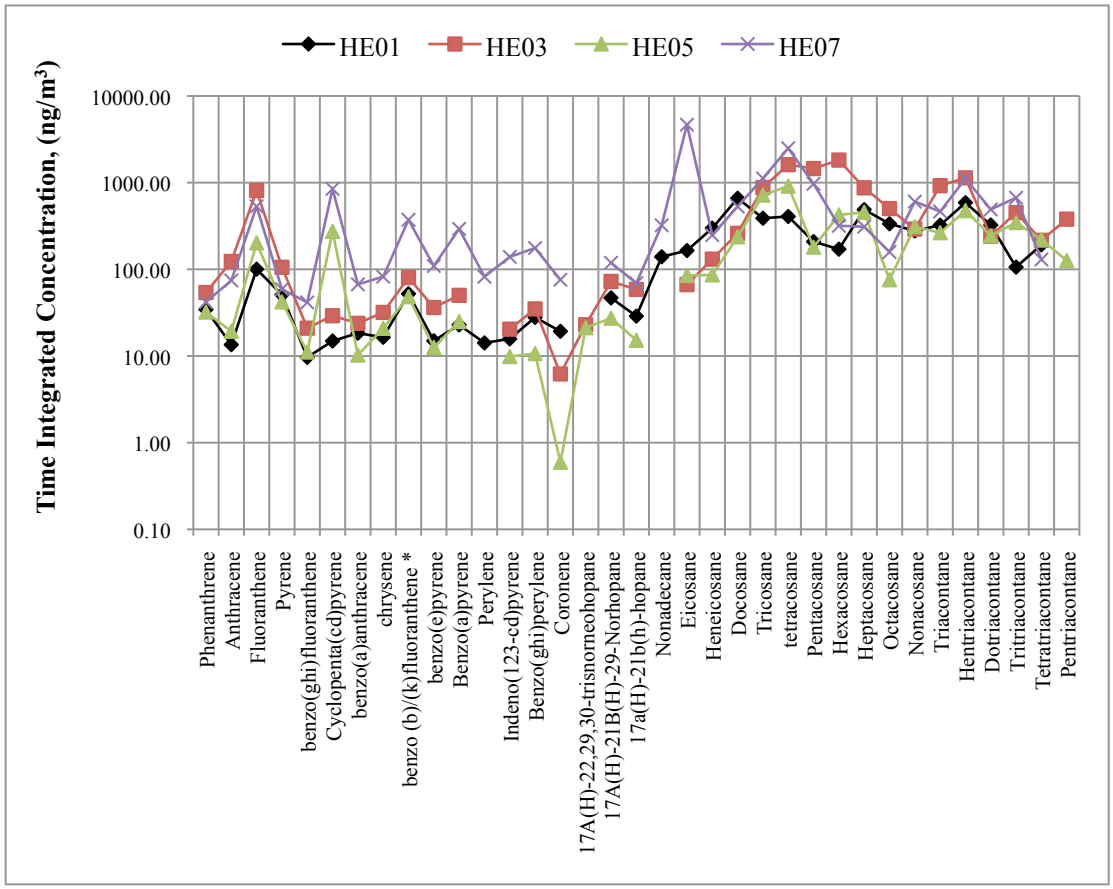

Figure 4.2: Organic compound concentrations

4.3 Real-time Measurements

The real-time measurements that have been analyzed from the ACE study include the AeroTrak and the MicroAethalometer. However, different than the time-integrated data, only three of the first four tests could be analyzed. The $PM_{2.5}$ volume concentration, and black carbon mass concentration data are presented in Table 4.5 for commutes HE03, HE05, and HE07.

Table 4.5: AeroTrak $PM_{2.5}$ volume concentration and MicroAeth black carbon mass concentration

		ACE-HE03	ACE-HE05	ACE-HE07
$PM_{2.5}$ (cm^3/m^3)	Average	4.90E-06	2.81E-06	5.74E-06
	Max	1.23E-05	4.48E-06	1.05E-05
	Min	1.92E-06	1.45E-06	3.26E-06
Black Carbon (ug/m^3)	Average	6.50	2.88	9.53
	Max	67.82	17.95	18.69
	Min	0.58	0.54	2.41

The values seen in Table 4.5 show that a rain event, which was encountered during the HE05 test, greatly affected the average concentrations and maximum concentrations. This is important in determining that the overall averages of commutes are representative of the larger scale events such as rain or other meteorological events. Further correlation is seen when approaching the data in the 5-minute averaged data seen in Figures 4.3, 4.4, and 4.5.

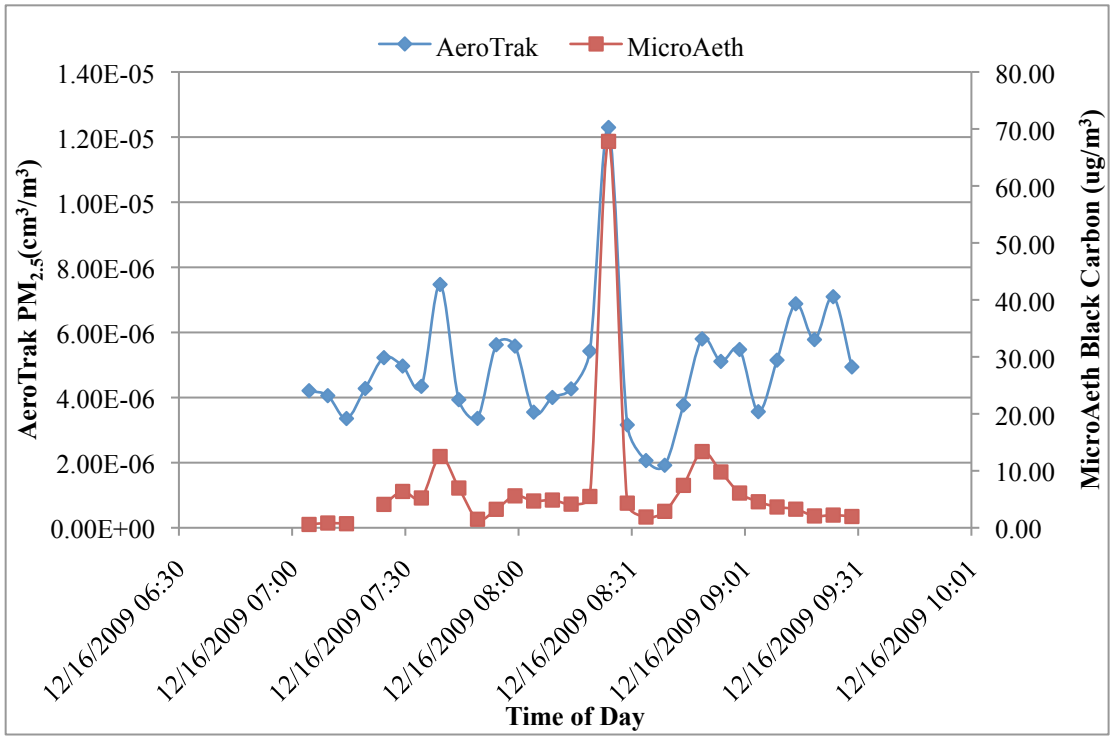

Figure 4.3: Real-time $PM_{2.5}$ volume concentration and Black Carbon concentration ACE-HE03

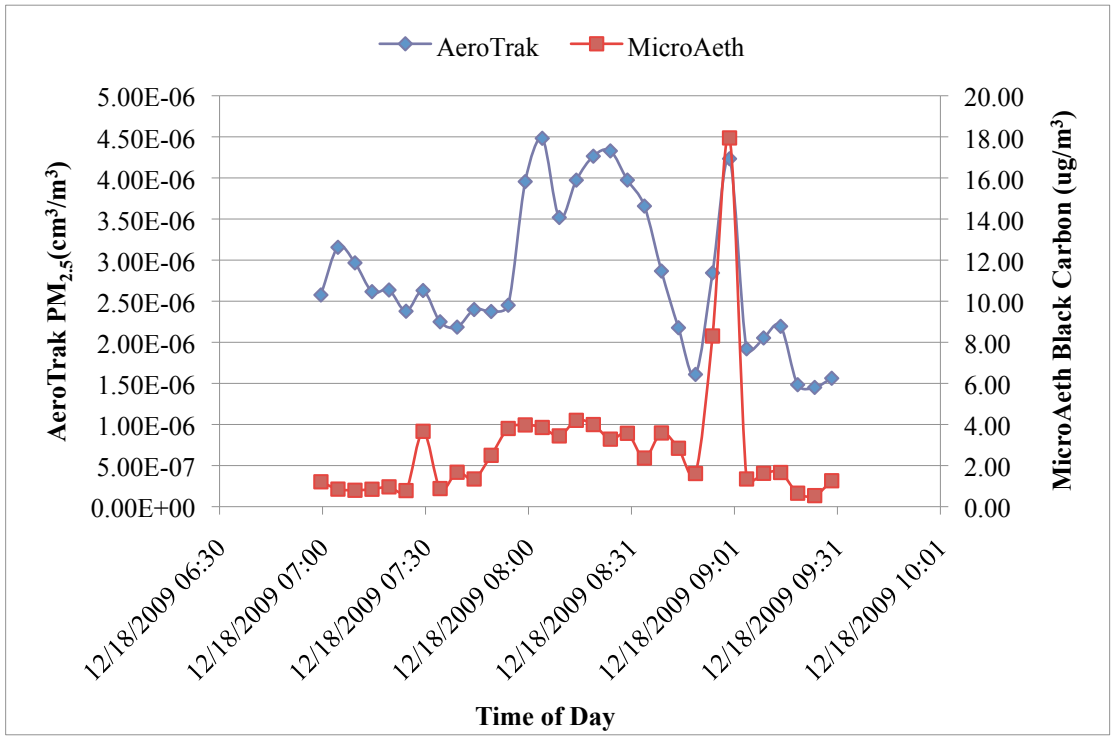

Figure 4.4: Real-time $PM_{2.5}$ volume concentration and Black Carbon concentration ACE-HE05

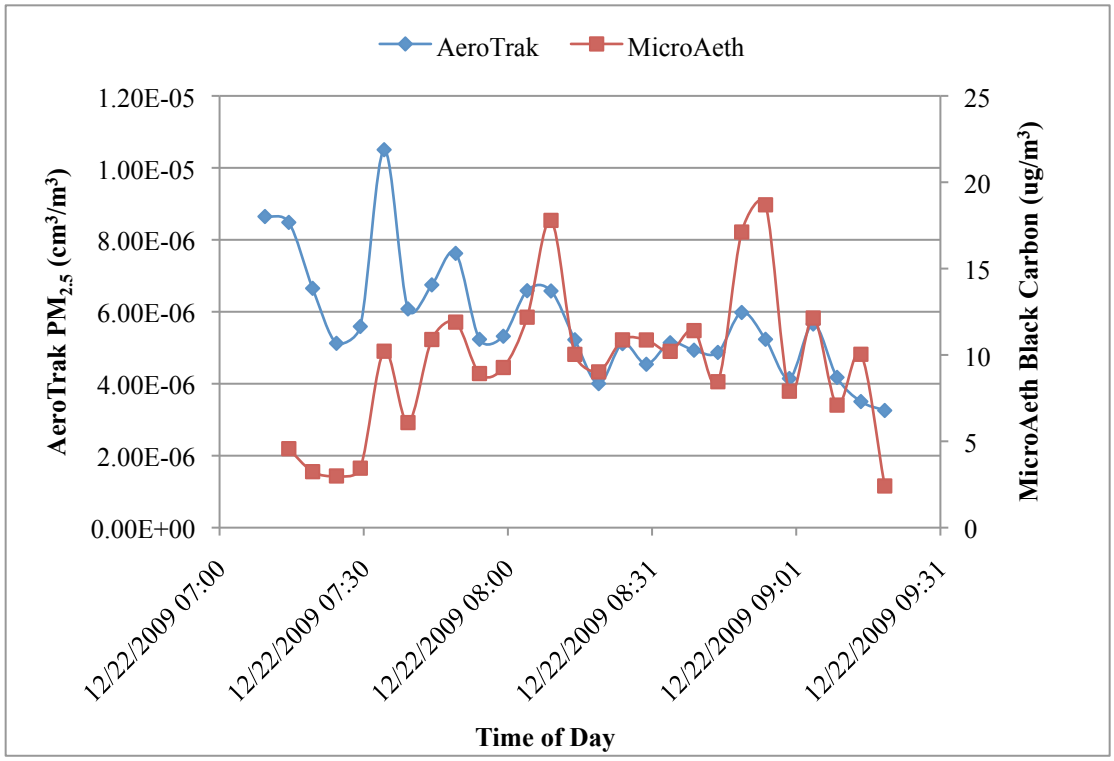

Figure 4.5: Real-time $PM_{2.5}$ volume concentration and Black Carbon concentration ACE-HE07

Even though the MicroAethalometer does not have a $PM_{2.5}$ cut off, the values seen in Figures 4.3, 4.4, and 4.5 show a high measurement association. When the AeroTrak values reached maximum peaks the MicroAethalometer reached peaks at the same time steps. Although this is seen, a relatively low correlation was calculate between the two instruments of r=.30. However, with the addition of more data points, as the ACE study moves forward, this value is expected to increase as outlying data points will not be as influential in the correlation calculation. Even though the magnitude was not the same because of the instrument sample sizes, it is seen that the generation of black carbon is also resulting in the generation of $PM_{2.5}$.

Another trend shown by these data is that the elevated values occur within 15 minutes of commutes being started and while the vehicles were moving. Also the data reaches maximums when the vehicles were located on the major highways of Atlanta (I-285 between I-75 and I-85) and not the service streets. This trend shows the dependence of PM concentration to on-road vehicle density. In addition, this shows that the particulate matter that is getting into the car is from outside sources and the air inside the car cabin is not being recirculated through the instruments, which was a main concern when developing the sampling package.

CHAPTER 5: CONCLUSION

This thesis presents preliminary results for the ACE study, a joint effort between the Georgia Institute of Technology and Emory University. The primary objective was to measure the concentration, size distribution and the chemical composition of $PM_{2.5}$ inside the vehicle cabin for several commuters. The in-vehicle exposure that is generated is an important aspect of daily human health. The data collected during a two-hour "in traffic" time limit for the fist four tests (HE01, HE03, HE05, HE07) provides a large enough time resolution for both time-integrated and real-time data measurements to be made. One of the main concerns of the ACE study was the time resolution needed for values of elements and organics to exceed the associated detection limits. The first four tests show that the detection limits for 19 elements are exceeded. Thus the use of Enrichment Factors in the elemental analysis shows the elements with EF greater than 1000, copper, zinc, barium, antimony, selenium, and sulfur, are associated with various to anthropogenic sources including brake wear, engine wear, and/or fossil fuel combustion.

In addition to the elemental species, 35 specific organic compounds were characterized in the ACE tests. Tetracosane, tricosane, pentacosane, hexacosane, eiocosane, hopane, and norhopane are all linked to mobile sources and were seen to have elevated concentrations in vehicle cabins. Also a trend of increasing organic concentration with increasing molecular weight was seen in all of the preliminary tests. The completion of the ACE study in the future should provide greater understanding specific organics associated with in cabin vehicle exposure.

The higher than normal values of the OC, EC, and WSOC measured during the ACE study shows above average exposure during a commute. The average ACE OC/EC ratio of 6.5 was lower than the Carrico et al. (2003) Atlanta Supersite Study, however, was within the range of typical urban OC/EC ratio of 4.5-9.4 determined by ASACA (Trail, 2010). The data shows that there are a high number of gasoline and diesel mobile sources, generally associated to OC and EC respectively, encountered during the commutes. This is seen through the elevated average OC and EC values, 36.1 $\mu g/m^3$ and 5.6 $\mu g/m^3$ respectively, which are not represented by the OC/EC ratio. The formation of WSOC does not increase by the same magnitude as shown by the WSOC/OC ratio of .17. This value is much lower than the Weber et al. (2007) highway WSOC/OC value of .44 and suggests that SOA is not influential in the elevated OC values.

The real-time instruments that were used show that there is sufficient circulation in the vehicle cabin to detect and resolve changing concentrations. The AeroTrak and the MicroAethalometer shows that the presence of black carbon are linked to $PM_{2.5}$. The use of these instruments with different measurement resolution can be averaged to the same time-scale to reinforce the same trends in maximums and minimums witnessed during the first ACE commutes. The real-time instruments that were used in the ACE study shows the data has high variability but overall averages of the data tend to agree.

In the future, more work will be completed in analysis of all of the time-integrated and real-time data. The purpose of this thesis was to create a portable package for the measurement and speciation of the in-vehicle ambient aerosols as

well as test methodologies for data analysis. It is seen that all of the measurements, both real-time and time-integrated, which were analyzed exceed detection limits. The goal of the portable sampling package was achieved given the small "in traffic" time scale. The future analysis will allow for greater understanding of exposure concentrations and in turn when compared to the health measurements made by Emory University it will allow for a more complete understanding of how human health is directly affected by exposure in traffic.

APPENDIX A: EXCLUSION CRITERIA FOR TEST SUBJECTS

Prospective participants who meet the following criteria will be excluded from participation in this study:

- have been diagnosed with diabetes;
- have ever in their lifetime suffered a myocardial infarction;
- have been outfitted with an implantable cardioverter-defibrillator or pacemaker;
- are currently taking digoxin or beta blockers for the treatment of hypertension or cardiac arrhythmias;
- have been diagnosed with a non-asthma pulmonary disease such as COPD, emphysema, or any type of lung cancer;
- have a forced expiratory volume in 1 second (FEV_1) less than 70% predicted at baseline;
- if asthmatic, cannot tolerate a bronchodilator withhold of 24 hours for long-acting drugs (salmeterol, formoterol, fluticasone/salmeterol) or 4 hours for short-acting drugs (albuterol, levalbuterol);
- smoke tobacco products or are residentially exposed to second-hand tobacco smoke (within the previous year) and
- do not possess a valid driver's license or have legal access to a properly insured vehicle.
- currently report that they are pregnant. (If participants become pregnant during the study, they will be asked to notify study personnel, and they will be removed from the study.)

APPENDIX B: SIEVERS WSOC CALIBRATION

Standards for Sievers 900 WSOC calibration were created using Oxcilic Acid Powder and then mixed with low TOC DI water. The resulting mixture was used to create 4 standard know solutions with 200 ppb, 300 ppb, 400 ppb, and 600 ppb of Water Soluble Organic Carbon. The resulting calibration curve was created and is seen in Figure B.1.

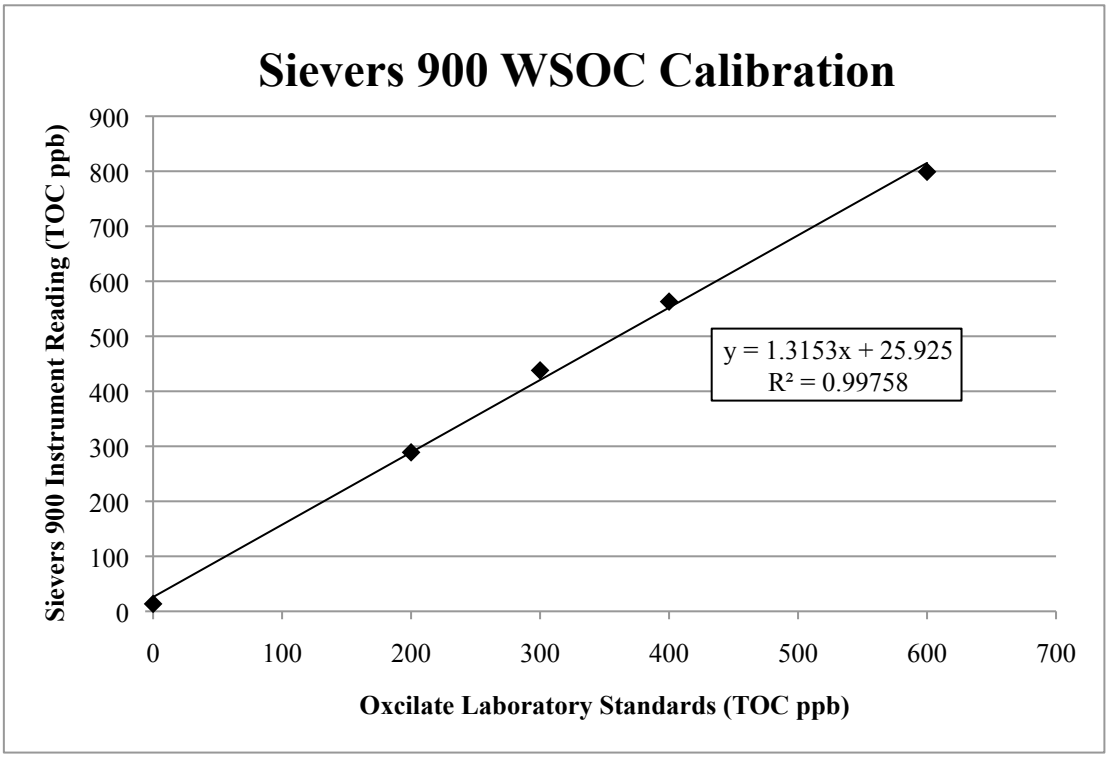

Figure B.1: Sievers 900 WSOC Calibration (June 4, 2010)

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