SPATIAL AND TEMPORAL TRENDS OF ORGANIC AND ELEMENTAL CARBON AS A COMPONENT OF $PM_{2.5}$ WITHIN THE NEW YORK METROPOLITAN AREA

by

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ABSTRACT OF THE THESIS

Spatial and Temporal Trends of Organic and Elemental Carbon as a Component of $PM_{2.5}$

Within the New York Metropolitan Area

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The goal of the project is to identify the spatial and seasonal patterns of Organic Carbon (OC), Elemental Carbon (EC) and fine particle mass ($PM_{2.5}$) in the NY City metropolitan area. This information is needed to assist with the development of State Implementation Plans (SIPs) for the control of fine particles in the NY City tri-state area and Mid-Atlantic Region, in order to meet EPA air quality standards. This study investigates the carbonaceous fraction of $PM_{2.5}$ in the NY City metropolitan area over an annual cycle from May 2002 to May 2003 to provide detailed analyses of the OC and EC carbon components and insights into their possible sources. Two sampling networks, the Speciation of Organics for Apportionment of $PM_{2.5}$ (SOAP) and the Speciation Trends Network (STN) provided separate measurements of OC and EC. It was found through linear regression analysis that the SOAP network sampling equipment measured OC and EC ambient mass concentrations values consistently lower than the STN EC and OC concentrations were compared between a heavily trafficked site, such as Elizabeth, NJ (NJ Turnpike, Toll Plaza 13), and a rural background site, Chester, NJ. Urban Queens, NY and suburban Westport, CT locations also were monitored as intermediate traffic sites with high to moderate population density. Time series data showed that all sites showed an OC and EC peak during the winter, while the Chester, NJ site had a peak of OC in the summer. The Chester site also had a high peak of the OC/EC ratio in the summer as well. From this, it was determined that motor vehicle traffic, a primary source of $PM_{2.5}$, EC and OC mass was found to dominate at the Elizabeth, NJ site, whereas at the rural low traffic Chester, NJ site, secondary OC mass showed greater input.

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Introduction

I. Particulate Matter and $PM_{2.5}$

Airborne particulate pollution has been one of the most prominent environmental concerns for major metropolitan areas worldwide. Particulate pollution is a conglomeration of suspended solid and liquid particles that are classified as “coarse” and “fine” particles. As particles are generally irregular in shape, these classifications refer to the “aerodynamic diameter” of a theoretical particle with a density of 1g/cm^3, but the same velocity and wind resistance of the irregularly shaped particle of interest (National Ambient Air Quality Standards 2007). Coarse particles, or PM_{10}, are classified as particles under 10 μm across in aerodynamic diameter, and are primarily attributed to suspended dusts, soils, and crustal materials from construction processes and to biogenic sources (Pope et al. 2006; NARSTO 2004; Simoneit and Mazurek 1982). Fine particles, or $PM_{2.5}$, are classified as particles under 2.5 μm in aerodynamic diameter, and have among their major sources combustion processes from industrial and motor vehicle sources (Mazurek 2002; Fraser et al. 1999; Kleeman et al. 2000; Rogge et al. 1993a; Schauer et al. 1999a). $PM_{2.5}$ in high concentrations has been attributed to short and long term exposure human health problems as well as a major contributor to urban smog and decreased visibility (USEPA 2004).

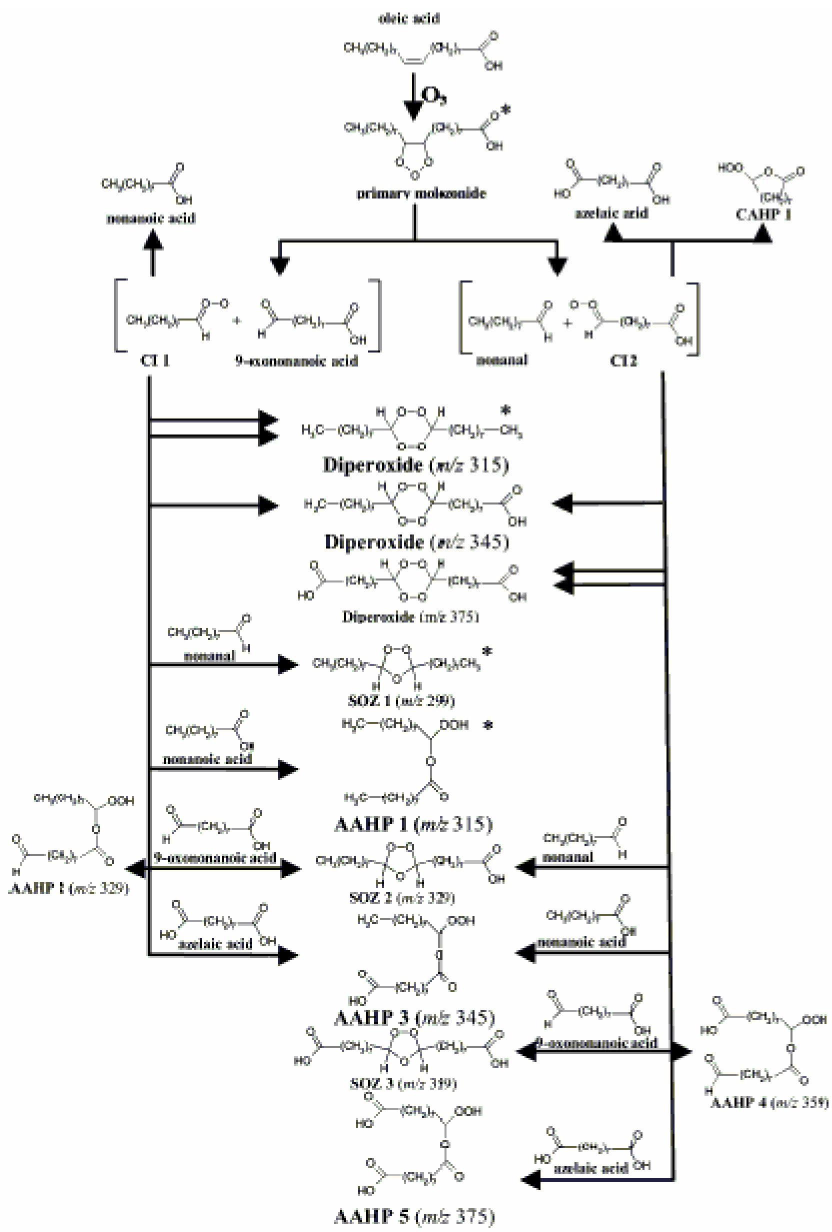
As part of the 1990 Clean Air Act, the United States Environmental Protection Agency, the USEPA, established a set of National Ambient Air Quality Standards (NAAQS), on which to regulate airborne contaminants nationwide to minimize negative effects on health and visibility. Currently, the New York metropolitan area is in non-attainment of $PM_{2.5}$, which is set at a regulatory level not to exceed 15 ug/m^3 per day

(USEPA 2004). Therefore, it is important to identify the sources of fine particles in the New York City area to develop effective control strategies that reduce emissions and lower ambient mass concentrations. This study was carried out to characterize the spatial and seasonal abundances of organic molecular markers, elemental carbon (EC), organic carbon (OC), and $PM_{2.5}$ in the New York area. In doing so, the goal was to determine the dominant sources of carbonaceous fine particles and to aid with the development of State Implementation Plans (SIPs) for $PM_{2.5}$.

In the Northeastern US $PM_{2.5}$ is generally made up of sulfates, nitrates, crustal elements, and carbon (NARSTO 2004). The carbon component is made up of both organic and elemental (often referred to as “black”) carbon. EC is associated with incomplete combustion reactions of organic molecules, such as the burning of fossil fuels. OC has a variety of sources, including primary sources, such as industrial and motor vehicle combustion. In addition, OC can have secondary sources such as the photochemical reaction products of atmospheric low molecular-weight organic compounds, and biogenic sources such as vegetation (Rogge et al. 1993a; Zheng et al. 2005). EC is almost exclusively contributed by incomplete combustion processes. OC has both primary and secondary sources, which are difficult to distinguish chemically by direct analytical methods (Chu 2004), thus indirect statistical methods are used.

Secondary organic aerosols (SOAs) result from the photochemical conversion of airborne organic molecules. Precursor organic molecules are emitted from a wide variety of primary sources that are manmade and natural. These are commonly low molecular-weight compounds, or VOCs, and are emitted by vegetation, mobile sources, and industrial and manufacturing sources. The precursor compounds have unsaturated carbon-carbon double bonds that are easily oxidized in the atmosphere. In the presence of sunlight, these organic molecules react with highly reactive ozone (O_3) molecules or radicals such as hydroxyl, peroxy, and alkyl radicals ($R\text{-}H^*$) (Atkinson et al. 2003). A sample reaction pathway is shown in Figure 1 below for oleic acid, a common molecular marker in urban fine particles (Rogge et al 1993c; Li et al 2006):

Figure 1- Sample Reaction Pathway of Oleic Acid with Ozone (taken from Reynolds et al. 2007)



The ability of alkyl radicals or ozone to attack numerous possible reactive carbon bonds results in a diversity of reaction products (Reynolds et al. 2006). In some instances a secondary organic molecule can be indistinguishable from the same compound emitted from a primary emission source since the only real difference between the two would be in the chemical formation of each. The impact of photochemical reactions in forming SOAs is quantified through the use of molecular tracer molecules with well-defined sources, and with the bulk carbonaceous components such as EC and OC which comprise the total fine PM mass (Hildemann et al 1994; Mazurek et al. 1997).

II. Scientific Approach

This study investigates OC, EC and $PM_{2.5}$ spatial and seasonal abundances during May 2002 to May 2003 at four monitoring sites located in the New York City area, encompassing Northern New Jersey, New York City, and southeastern Connecticut. The four sites were selected based on the expected level of urban emission sources within the metropolitan area, proximity to heavily traveled roadways, and prevailing wind direction in terms of "upwind" or "downwind" of NY City. All sites have $PM_{2.5}$ mass comprised of EC, OC and inorganic compounds plus sorbed water vapor (Rogge et al. 1993c). Atmospheric OC $PM_{2.5}$ mass can be contributed as secondary carbon from the photochemical oxidation of manmade and biogenic emission (Rogge et al. 1993a). However, EC does not have a photochemical origin and is contributed only from incomplete combustion processes by primary emission sources (motor vehicles, construction equipment, wood and biomass combustion, industrial emissions). Both OC and EC are present in some proportion in primary sources (Rogge et al. 1993c). It is not known how the secondary OC and primary OC and EC mass vary spatially and seasonally throughout the NY City area. The mass balance of OC and EC to total $PM_{2.5}$ mass is one approach that can track the variation and provide evidence of primary versus secondary input. Therefore, daily $PM_{2.5}$ samples collected over an annual cycle throughout the metropolitan NY City area are necessary to evaluate the changing mass balances seasonally and temporally.

Two collocated fine particle sampling networks provided an entire annual cycle of OC, EC and $PM_{2.5}$ ambient mass measurements for this study: the US EPA Speciation Trends Network (STN) and the Speciation of Organics for the Apportionment of $PM_{2.5}$ project (SOAP). The SOAP experiment was a collaborative study by Drexel and Rutgers Universities. It was supported by the Northeast States Coordinated Air Use Management (NESCAUM), and involved field sites operated by the New York State Department of Environmental Conservation (NYSDEC), the New Jersey Department of Environmental Protection (NJDEP), and the Connecticut Department of Environmental Protection (McDow et al. 2007). The four SOAP monitoring sites were collocated with the STN sites to allow access to other gas and particle measurements and meteorological data on sampling days. The SOAP and STN networks collected daily fine particle samples on the one-in-three day STN schedule. $PM_{2.5}$ mass was obtained from a separate STN network instrument operating at each site. These data provided the total fine particle masses used in the mass balance calculations. However, the STN and SOAP networks had different samplers with different flow rates and exposed filter areas to collect fine particles. Sample filters from both networks used the same analytical method to determine OC and EC mass loadings. In order to compare ambient concentrations of

OC and EC, it is first necessary to examine the blank levels from both networks in order to establish possible sampling and analysis bias. Following this analysis, the second task in this research project will use the collected $PM_{2.5}$ data to quantify the difference, if any, between the SOAP and STN measurements.

A third task is to generate an understanding of primary versus secondary sources of organic carbon. This will be accomplished by analysis of the seasonal and spatial trends of the chemical mass balances derived from task two and the relationship of OC to EC concentrations (OC/EC ratios) at the four sites. Gray et al (1984) identified EC as a tracer of the aggregate primary emissions to urban, suburban and rural air sheds. This was based on an intensive study of the chemical mass balance for fine particle mass, OC, EC and inorganic compounds over an annual cycle in the Los Angeles air basin. The ratio of the ambient mass concentrations of OC to EC (OC/EC) tracks the variation over space and time and is not dependent on the height of the surface mixed layer. The depth of the mixed layer varies seasonally and spatially, thus influencing the absolute concentrations of OC, EC and $PM_{2.5}$ mass (Gray et al. 1984; Rogge et al 1993c; Turpin et al 1991, 1995). A 2003 study by Gioia and coworkers of OC/EC ratios in $PM_{2.5}$ mass sampled from rural and urban NJ sites showed seasonal and spatial differences. Higher levels of secondary OC were estimated at the rural sites relative to urban sites. Also, higher levels of secondary OC occurred during summer months compared to colder months. The experimental design of the study used filters obtained only for specific seasons and not over an annual cycle with a consistent 1-in-3 day sampling schedule.

As an alternative approach consistent with the approach of Gray et al 1984, this thesis research will examine OC/EC ratios for the SOAP and STN network filters over an annual cycle that provide periods of high proportion (summer – high photochemical activity) and low proportion (winter – low photochemical activity). The difference in OC/EC ratios comparing summer versus winter values can be used to approximate the contribution of secondary OC to the total OC mass in the NY City metropolitan area. As part of the third task the trends in OC/EC ratios will be analyzed over the course of the sampling study at the four SOAP/STN sites. It is hypothesized the lowest variation in OC/EC will be at those sites dominated by primary emission sources. In addition, the highest proportion of secondary OC is expected to be present at the upwind and downwind sites. This is due to the greater proportion of regional background $PM_{2.5}$ mass and to decreased urbanization resulting in fewer local primary sources.

III. Scientific Questions

This work is an investigation of three essential questions regarding $PM_{2.5}$ and its carbonaceous fraction in the New York City Metropolitan area. The first question examines the trends of OC and EC fine particle mass: How do concentrations vary according to space and time? This analysis will be accomplished by comparing both daily and seasonally averaged ambient concentrations of OC and EC, and by using box and whisker plots generated by the mathematical software Matlab 7.4. Secondly, how do the SOAP and STN EC and OC ambient concentrations differ when analyzed using statistical analyses such as the F-test, and linear least squares regression analysis? Thirdly, what insights can be gained from the data about the influence of secondary organic carbon concentrations? The relationship between OC EC mass and an estimate of secondary OC mass will be determined by tracking trends in OC/EC ratios.

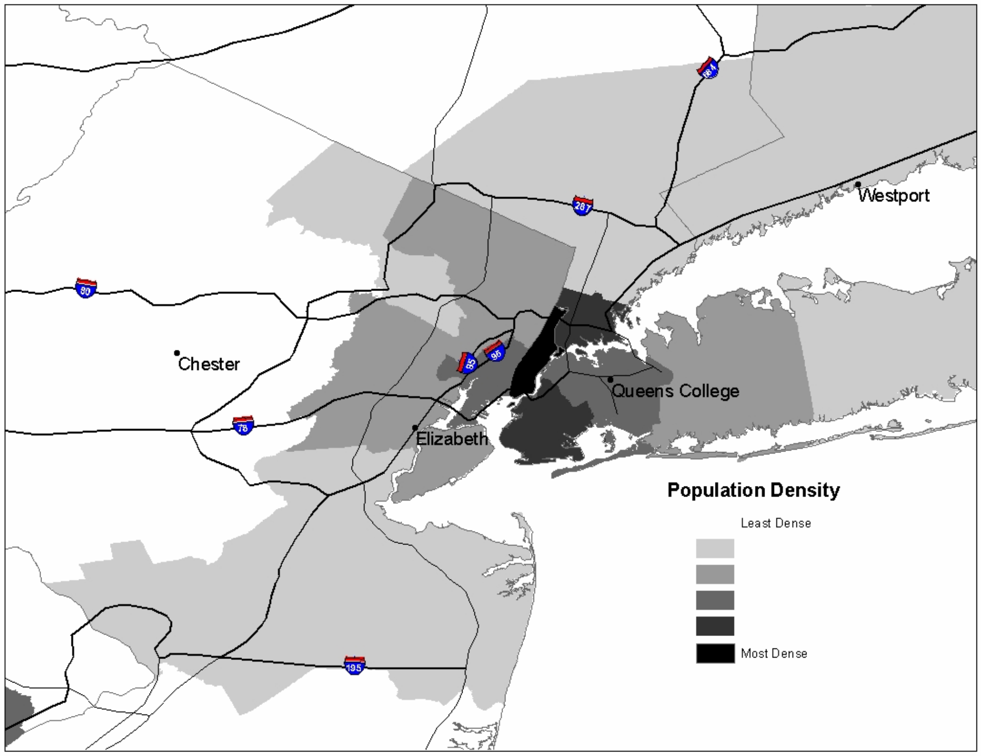
Experimental Methods

I. Sampling Network:

Fine particle samples (nominal aerodynamic particle size < 2.5 µm, PM2.5) were collected by the Speciation of Organics for Apportionment of $PM_{2.5}$ (SOAP) Study in the New York, New Jersey, and Connecticut area. The aim of the project was to track organic molecular markers that were indicative of a particular emission source of OC. A full description of the SOAP methods is reported by McDow et al., 2007. The SOAP network operated from May, 2002 to May, 2003 at four air quality monitoring sites, collocated with the EPA Speciation Trends Network (STN) network for $PM_{2.5}$. The collocation of the two sampling studies allows for a comparison of the organic, elemental, and total carbon measurements reported by the SOAP and STN networks.

The first monitoring site was in Queens, NY and was the principal EPA Supersite monitoring station at Queens College in New York City. Upwind of this site was the Elizabeth, NJ site located adjacent to the NJ Turnpike Exit Toll Plaza 13. The NJ Turnpike site is expected to be dominated by transportation emissions including those from the New Jersey Turnpike, the Port of Elizabeth, and Newark Liberty International Airport. Downwind of the Queens Supersite was the Westport, CT monitoring site located in a suburban area in Fairfield, CT. The final site was a regional background site in Chester, NJ, and predominantly upwind of the NYC metropolitan area. The Chester, NJ, site was considered to be a regional background site for the metropolitan NYC air shed (McDow et al. 2007).

Figure 2- Population Density Map of SOAP/STN Study Area (McDow et al. 2007)



The SOAP monitoring sites, shown in the map in Figure 1, were selected to evaluate the fine particle concentrations in relation to urban emission sources, including various degrees of on-road mobile sources from diesel and gasoline-powered motor vehicles. The proportion of secondary fine particle mass is expected to vary throughout the study site with the greatest proportion at the Chester NJ upwind site. The Queens College site is representative of a dense urban population served by heavily used highways. The Elizabeth, NJ site is situated outside of Toll Plaza 13 of the New Jersey Turnpike, where an estimated 220,000 motor vehicles pass through on a daily basis (Ozbay 2006). The Elizabeth, NJ is within close proximity (0.8 miles) of the Newark Liberty International airport, with its significant air traffic. The Westport, CT site is in a suburban area with modest traffic, but located in a county where four major power plants each emit more than 20 tons of PM2.5 annually. The Chester, NJ monitoring site is in a rural area with low traffic, upwind of the NYC metropolitan area, and is a low population density residential area with large areas of undeveloped land. Also, shown in Figure 1 is the relative population density for the SOAP and STN sampling sites. The highest population density is in Manhattan and Queens, NY and lowest is the Chester, NJ site.

II. Samplers:

Daily PM2.5 samples were collected from May 26th, 2002 to May 30th, 2003. The SOAP network used new TISCH 2 or 4-channel samplers to collect the PM2.5 samples. The fine particles were filtered from the ambient air stream with pre-baked quartz fiber filters. Samples were collected according to the STN sampling schedule, which was once every 3 days. Filters were collected over 24 hours from midnight to midnight. By collecting the SOAP samples on the same day as the STN samples it was possible to directly compare STN and SOAP ambient PM2.5 elemental (EC) and organic carbon (OC) concentrations. It also is possible to perform statistical analysis on the similarity of the two measurements at each monitoring site.

Overall the number of successful daily filters collected shows that the SOAP sample collection was much more complete (ambient concentrations given in appendices) than the STN study as SOAP had more reported EC and OC mass concentrations over the course of the year. The subsequent statistical analysis of the ambient EC and OC mass concentrations take into consideration the different number of ambient filters (SOAP& STN daily pairs) at each site, which totaled to 226 matched pairs of SOAP and STN filters. In total, over 700 filters were successfully collected by the SOAP network across the four monitoring sites. Seasonal composites were compiled from this total number of filters as a subgroup of SOAP daily filters. The seasonal composites were generated based on the requirement that all sites had a successful filter for a given season. The exception was the early summer 2002 period which had successful filters collected only at the Chester, Elizabeth, and Queens sites. Flow control problems at the Westport site were corrected by mid-summer 2002.

The SOAP PM sampling protocol was designed based on the collection methods of the STN program. One major difference between the two was the $PM_{2.5}$ samplers used at the four sites. The SOAP study used the Tisch Environmental TE-1202 and TE-1204 samplers equipped with 2 and 4 flow channels, respectively. The Tisch samplers were fitted with Teflon gaskets as to reduce organic background contributed by the sampling environment. Flow rates for the Tisch samplers were 113 liters per minute. The STN employed the Spiral Aerosol Speciation Sampler (SASS; Met One Instruments, Grants Pass, Oregon). The flow rate for the SASS samplers was 6.7 liters per minute (USEPA 2000). Both sampling networks collected particulate samples on quartz fiber filters (SOAP 102 mm diameter; STN 47 mm diameter). The STN filters were baked at 850 °C and the SOAP filters at 550 °C to remove organic contaminants. The SOAP filters were processed in pre-baked individual aluminum envelopes and were removed from the envelopes just before loading into the filter holder cassettes.

Field, dynamic, and trip blanks were collected as routine protocol throughout the SOAP network operation. Comparable values for the STN blanks were not available for comparison to the SOAP blanks. It was necessary to estimate the STN OC and EC blanks via a linear regression method against $PM_{2.5}$ mass. A discussion of estimated blank values for STN will be made below. The SOAP blanks were analyzed for OC and

EC mass using the same protocol for the ambient samples. Field blanks were placed in the Tisch Sampler without air flow. Dynamic blanks were placed in the sampler and exposed to air flow for 10 minutes. Trip, or travel, blanks were not subjected to any airflow, but simply were transported and stored the same as the ambient sample filters (McDow et al 2007).

III. Carbon Analysis:

The mass of EC and OC on each SOAP daily filter was measured from a 1 cm^2 filter punch that was removed and analyzed separately. The punch equaled 1.76 % area of the total exposed filter area (56.72 cm^2). The volume of airflow through each SOAP filter was 163 m^3 per day. The filter punches were sent to Sunset Laboratory for EC, OC and total carbon (TC) analysis by the NIOSH 5040 method (Peterson et al. 2002). Ambient mass concentrations for EC and OC were reported by Sunset Labs in units of mass in micrograms (ug) per square centimeter (cm^2). Mass concentrations in micrograms per square meter (ug/m^3) were calculated by multiplying the reported EC and OC mass of the aliquot by the total exposed area and dividing by the total volume of air sampled:

$$[\text{Ambient Concentration}] = [\text{Aliquot Mass } (ug/cm^2)] \times [\text{ Total Exposed Area } (cm^2) / \text{Total Air Volume } (m^3)].$$

The NIOSH Method 5040 bulk carbon analysis method has been critically reviewed and compared to other thermo-optical methods by Chow et al (2001) and Bae et al. (2004). The basis of each method involves operationally defined temperatures with separate pyrolysis and combustion steps. The NIOSH Method 5040 temperature protocol used by the Sunset Laboratories EC/OC Carbon Analyzer is listed in Table 1. A brief overview of the method follows.

The 1 cm^2 filter punch was treated to four consecutive temperature increases in an oxygen-free, high purity helium atmosphere. This temperature elevation without oxygen results in the pyrolysis of some organic compounds to elemental carbon (EC), which was monitored by laser beam transmission and then corrected for the added EC as part of the post-run data analysis. During the first heating step, some organic compounds were vaporized to CO_2, which was reacted to form methane, CH_4, and then monitored by a Flame Ionization Detector (FID).

Table 1- NIOSH 5040 EC/OC Determination Temperature Protocol (Sunset Labs)

Atmosphere	Time (s)	Temperature (^{o}C)
Helium	60	310
Helium	60	475
Helium	60	615
Helium	90	870
Helium	cool down step	35
Helium/Oxygen	45	550
Helium/Oxygen	45	625
Helium/Oxygen	45	700
Helium/Oxygen	45	775
Helium/Oxygen	45	850
Helium/Oxygen	120	890
Calibration (CH_4)	110	-

After the oxygen-free step, the analyzer oven was cooled and the pure helium gas feed was switched to a 2% oxygen/helium mixture. Subsequently, the temperature was raised incrementally to 900 °C. During this step the original EC and the EC formed in the first pyrolysis step were combusted to form CO_2. The CO_2 was converted to CH_4, methane, by the methanator, and once again was monitored by the FID. Once carbon is no longer being oxidized, an internal standard of methane gas is injected into the oven.

This allows for an internal reference of previously calibrated methane gas concentration to be established. The NIOSH method operationally defines that prior to the point in the second heating step where the laser beam transmission returned to the original sample transmission, the instrument is responding to OC, and after that point, the apparatus is measuring EC. Replicates, instrument blanks and calibration data were obtained by Sunset Labs and reported along with the ambient filter analyses.

Results and Discussion

I. Blank Estimation:

For the SOAP network, both field and trip blank samples were collected and processed to determine EC and OC concentrations. The calculations for average blank values are presented in Table 2. As reported by Sunset Laboratories, the reported uncertainty of the instrument was 6% of each ambient sample filter measurement. It should be noted that the reported OC and EC concentrations were in units of micrograms (μg) per square centimeter (cm^2). The reported OC and EC masses are integrated over the filter exposed area and assume loadings are uniform across. The OC and EC total mass loadings per filter were converted to an ambient concentration for the entire volume of air passed through the filter. The total air volume sampled by each SOAP filter was 162.72 m^3, per 24-hour period. Blank values for the SOAP filters were calculated from the reported instrument uncertainty of ±0.1 ug/cm^2. Therefore, the OC blank uncertainty was determined as follows:

$$0.06 \times \textit{Filter Conc. } (ug/cm^2) \pm 0.1\ ug/cm^2 \times (56.72\ cm^2 / 162.72\ m^3) = \textit{Uncertainty } (ug/m^3)$$

where 56.72 cm^2 is the total filter area and 162.72 m^3 is the total air volume. Based on the above equation, the blank value for OC in the SOAP network filters were estimated in the range of -0.01 to 0.06 ug/m^3, while the EC blank levels 0 ug/m^3, or otherwise below detection limits.

Because the STN did not report blank concentrations with its ambient concentration data, blank values were estimated from linear least squares regressions of the ambient OC, EC and $PM_{2.5}$ masses. The blank values for STN OC and EC samples collected during the SOAP network period (May 2002 to May 2003) were obtained by taking the y-intercept of the linear least squares regression line of ambient OC and EC mass concentrations against corresponding ambient PM2.5 mass concentration values. In linear least squares regression, a scatter-plot of the data points of the ambient OC or EC is plotted along with the corresponding $PM_{2.5}$ concentration for a given date. A straight line then was fitted to the scatter-plot in order to produce the straight line which best models the data, with ambient OC and EC data given as a function of $PM_{2.5}$ concentration. The criterion for the fitted regression line is that the square of the difference between the predicted species concentration and the reported species concentration be minimized. Thus, when $PM_{2.5}$ mass is zero, the mass of OC and EC also should be zero. A positive y-intercept using the ambient concentrations suggests possible systematic contributions of OC or EC mass to the sample filter. The regression lines were calculated in the software applications Microsoft Excel and Matlab 7.4, with the special curve fitting toolbox being used in the latter. In order for the regression line to be determined, only those ambient data points for OC and EC which had corresponding $PM_{2.5}$ measurements were used, as a scatter-plot otherwise could not be generated.

Table 2- Calculation of SOAP Ambient EC and OC Blank Values from Sunset Labs Analytical Data

	OC(ug/sq cm)	EC(ug/sq cm)	OC (ug/m3)	EC (ug/m3)
Chester	1.97	0.00	0.69	0.00
	1.76	0.04	0.61	0.01
	1.17	0.00	0.41	0.00
	1.16	0.00	0.40	0.00
Elizabeth	0.93	0.00	0.33	0.00
	1.40	0.00	0.49	0.00
	1.08	0.00	0.38	0.00
	Trip Blanks			
	0.60	0.00	0.21	0.00
	0.82	0.00	0.28	0.00
Queens	0.99	0.00	0.35	0.00
	0.97	0.00	0.34	0.00
	Trip Blanks			
	1.12	0.00	0.39	0.00
	0.96	0.00	0.33	0.00
Westport	3.00	0.00	1.04	0.00
	0.97	0.00	0.34	0.00
Average	1.26	0.00	**OC (ug/m3)**	**EC (ug/m3)**
Measured Unc. (+)	0.18	0.00	0.06	0.00
Measured Unc. (-)	-0.02	0.00	-0.01	0.00

The study by Kim et al. (2006) used regressional analysis blank calculations at three of the STN sites (Chester, Elizabeth, Queens) used in the SOAP/STN study. The Westport, CT site monitoring was included as a monitoring site at a later date than the other sites. Table 3 compares the calculations of regressed blank concentrations from the current study with those calculated in the Kim et al study. It is seen that the two regression calculations, spanning distinct but overlapping time intervals, yielded similar blank values. The difference shown by the two regressed blank calculations at Chester and Elizabeth is attributed to a combination of the different time intervals which monitored two different air masses with different primary and secondary emissions from local and regional sources. One also can observe from the table below that the intercepts calculated in the Kim study are within the 95% confidence bounds of the current calculated regression intercept, the boundaries in which there is a 95% certainty that the idealized statistical value would occur. The similarity of the STN blank estimations using the regression approach is significant since it supports the case for similar bias contributed by sample collection, handling, and analysis and to very slight changes in the variation of ambient mass concentrations of OC and EC for the two STN sampling periods. If vastly different natural ambient conditions were the dominant influence, then the slope and y-intercept of the modeled data would be different for the Kim et al and SOAP STN regressed OC, EC and $PM_{2.5}$ mass concentrations.

Table 3- Comparison of STN OC Blank Approximations from Linear Least Squares Regression of Ambient OC Concentration versus Total PM2.5 Concentration

STN OC Blank Approximations (ug/m^3)

Site		Sampling Period	Intercept
Chester	Kim et al 2006	1/2002-12/2002	1.35
	Current Study	5/2002-5/2003	0.8391
		95% Conf. Interval	(0.2249, 1.453)
Elizabeth	Kim et al 2006	1/2002-12/2002	2.19
	Current Study	5/2002-5/2003	2.664
		95% Conf. Interval	(1.766, 3.562)
Queens	Kim et al 2006	4/2001-12/2003	1.57
	Current Study	5/2002-5/2003	1.638
		95% Conf. Interval	(1.02, 2.329)

The estimated blank values for the May 2002-2003 STN filters are reported in Table 4 and correspond to the y-intercepts of the regression lines. The values are interpreted as the estimated OC or EC mass concentration present on the ambient filters when there is no $PM_{2.5}$ concentration measured. The boundaries of the 95% confidence interval are listed along with the R^2 value. The regression lines have very low R^2 values (where a perfect fitting of the data would thus produce $R^2=1$), indicating that the regression lines determined were not well-fitted to the scatter-plots of the data. $PM_{2.5}$ mass alone, therefore, is not a good predictor of OC and EC mass. Other factors such as changing atmospheric conditions, seasonal differences in emissions sources, and systematic bias from sample handling, collection and analysis all combine to contribute to the uncertainty in the y-value as predicted by $PM_{2.5}$ mass.

Table 4- Calculated STN Blank Values and Confidence Intervals from Regression of Ambient EC and OC Concentrations with PM2.5

Speciation Trends Network (2002-2003)

Sampler Intercepts as Determined by Linear Least Squares Regression of OC,EC vs $PM_{2.5}$ Conc.

Chester					
OC (ug/m3)	**95% CI**	**R^2**	**EC (ug/m3)**	**95% CI**	**R^2**
0.8391	(0.2249, 1.453)	0.5927	0.2464	(0.1282, 0.3647)	0.2557
Elizabeth					
OC (ug/m3)	**95% CI**	**R^2**	**EC (ug/m3)**	**95% CI**	**R^2**
2.664	(1.766, 3.562)	0.5324	0.7034	(0.2638, 1.143)	0.3281
Queens					
OC (ug/m3)	**95% CI**	**R^2**	**EC (ug/m3)**	**95% CI**	**R^2**
1.675	(1.02, 2.329)	0.3127	0.4628	(0.2644, 0.6613)	0.1447
Westport					
OC (ug/m3)	**95% CI**	**R^2**	**EC (ug/m3)**	**95% CI**	**R^2**
0.7215	(0.1301, 1.313)	0.7376	0.08251	(-0.1328, 0.2979)	0.5572

Figure 3a illustrates a scatter plot of OC versus $PM_{2.5}$ ambient mass concentrations for STN ambient samples collected at Elizabeth, NJ during the SOAP operating period. The y-intercept is 2.44 $\mu g/m^3$ with a slope of 0.20. The scatter plot of OC versus $PM_{2.5}$ ambient mass concentrations from the SOAP filters collected with the Tisch 2-channel sampler over the same time period has a y-intercept of 1.49 $\mu g/m^3$ and slope of 0.19. Comparing the OC y-intercept of the SOAP blanks to the measured blank average of travel, field and dynamic blanks in Table 2, the method of regression overestimates the OC ambient blank by 1.4 to 1.5 $\mu g/m^3$. However, the difference in the y-intercepts between is about 1 $\mu g/m^3$ lower for the SOAP OC versus $PM_{2.5}$ scatter plot. Similarly, comparing the SOAP and STN y-intercepts for OC versus $PM_{2.5}$ mass demonstrated no consistent systematic trends in the difference between the SOAP and STN regression results. Queens had y-intercepts of 1.58 (SOAP) and 1.67 (STN) $\mu g/m^3$; Westport with y-intercepts of 1.21 (SOAP) and 0.72 (STN) $\mu g/m^3$; and Chester y-intercept values of 1.27 (SOAP) and 0.84 (STN) $\mu g/m^3$. Although the blank concentrations determined by regression for STN as listed in Table 4 were considerable, these most likely reflect the variability of ambient OC as a component of $PM_{2.5}$ mass rather than a systematic bias due to sampling, handling and analysis methods.

Figure 3a- Sample Regression Plot Regressing STN Elizabeth OC Concentrations

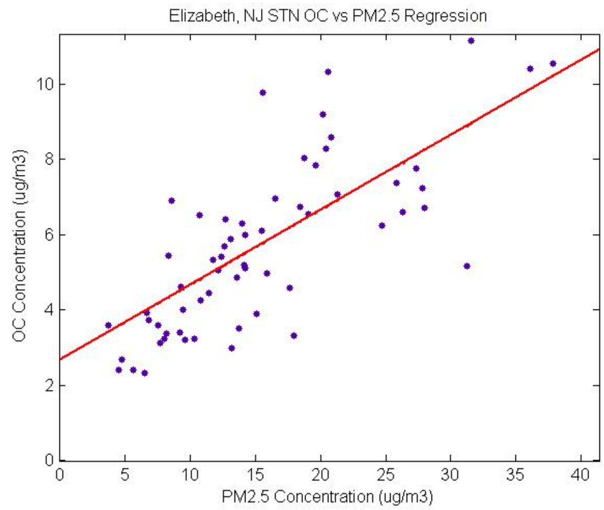
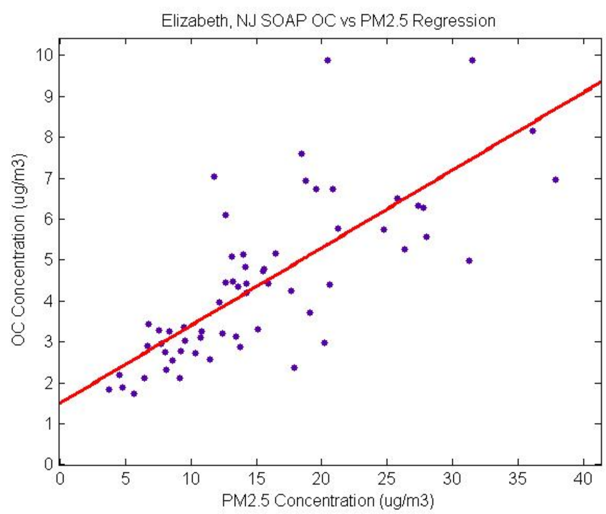


Figure 3b- Sample Regression Plot Regressing SOAP Elizabeth OC Concentrations



From the sample regression plot and STN ambient data shown in Figure 3a, the approximated STN blank values for organic carbon at the four sites are 25%, 45%, 51%, and 21% of the average yearlong OC concentrations at Chester, Elizabeth, Queens, and Westport respectively. For the Queens, NY site, though, the difference in both the mean and median values is negligible. This indicates that the regression of OC and EC ambient concentrations with $PM_{2.5}$ concentrations may not be an accurate method of calculating approximate blank concentrations. As mentioned above the variability in the data as shown by the low R^2 values indicates the influence of varying air mass trajectories and atmospheric conditions. The calculated STN blank values should be interpreted as estimates based on weakly correlated OC and $PM_{2.5}$ ambient mass concentrations.

II. Sampler Comparison:

A. Mean/Median Values:

Table 5 lists the mean and median data for both OC and EC 24-hour daily samples at each site. The data were averaged over the course of the year for all available reported filter data for the given species, as given in the Appendices. Table 5 shows yearly averaged concentrations for both studies of OC and EC. The measured ambient concentrations were consistently higher on average for the STN protocol for all the measurements except the Queens, NY, OC measurements. The EC mass concentrations are consistently lower than the OC mass concentrations at all sites for all days of the 2002-2003 annual cycle. The relative amount of OC in proportion to EC will be discussed later in this paper to address the contribution of secondary organic aerosol mass to $PM_{2.5}$. Generally, it is shown that the reported SOAP study average concentrations measured by the Tisch Environmental sampler are lower than STN averages obtained by the Met-One SASS, with smaller standard deviations, that is, smaller variation in the data set. The exception to this is the OC concentrations at the Queens monitoring site, though the median concentrations of OC measured at Queens were in agreement to the hundredths decimal place.

Table 5- Comparison of Selected Descriptive Statistics for SOAP and STN EC and OC Ambient Mass Concentrations

Average OC and EC Statistics from May 2002 - May2003

Chester, NJ	N	Mean	Median	StanDev	Minimum	Maximum	Range
SOAP OC (ug/m3)	78	2.6102781	2.12032962	1.36525455	0.737322404	7.602068807	6.8647464
STN OC (ug/m3)	58	3.31968394	2.76236108	1.83030414	0.974562973	9.932332454	8.95776948
SOAP EC (ug/m3)	78	0.2211114	0.16880784	0.30528931	BDL	2.605182799	2.6051828
STN EC (ug/m3)	58	0.4201273	0.38571453	0.18942419	0.113598506	1.040201521	0.92660302

Elizabeth, NJ	N	Mean	Median	StanDev	Minimum	Maximum	Range
SOAP OC (ug/m3)	76	4.62808053	4.3451507	2.087738	1.743758743	11.22522708	9.48146834
STN OC (ug/m3)	64	5.93088761	5.56970243	2.51089038	2.321157621	12.62152961	10.300372
SOAP EC (ug/m3)	76	1.26850821	1.09846762	0.81868022	0.112651641	3.776461545	3.6638099
STN EC (ug/m3)	64	1.75542925	1.49138882	1.0267358	0.024997516	4.651415965	4.62641845

Queens, NY	N	Mean	Median	StanDev	Minimum	Maximum	Range
SOAP OC (ug/m3)	77	3.54106273	3.12778198	1.49837675	1.216165866	7.8952387	6.67907283
STN OC (ug/m3)	67	3.25368672	3.14835308	1.48542556	0.403784699	7.917641657	7.51385696
SOAP EC (ug/m3)	77	0.62632686	0.53126521	0.36904498	0.105555731	2.038175752	1.93262002
STN EC (ug/m3)	67	0.74489775	0.6433879	0.40032665	0.09818	2.170010046	2.07183005

Westport, CT	N	Mean	Median	StanDev	Minimum	Maximum	Range
SOAP OC (ug/m3)	70	3.16316333	2.89685514	1.43233323	1.151955765	6.75056173	5.59860597
STN OC (ug/m3)	39	3.41943883	3.21645892	1.48423349	1.236390273	7.320362179	6.08397191
SOAP EC (ug/m3)	70	0.38269755	0.32441524	0.26176953	BDL	1.167331404	1.1673314
STN EC (ug/m3)	39	0.7398224	0.68199428	0.41604199	0.251409503	2.084159628	1.83275013

One-way analysis of variance (ANOVA) tests were performed over the paired STN and SOAP data sets at each site. In one-way ANOVA, the means of the two data sets are compared based on differences within one sample group compared to the sample mean, versus the observed differences between two sample means, in order to produce the F-statistic (a ratio of the sum of the squared differences among separate groups over the sum of the squared differences within groups). This analysis was performed with the Matlab-7.4 statistical toolbox. With a significance level of α=0.025, it was determined that the mean values of the two sample sets at Chester, NJ, Elizabeth, NJ, along with the EC sample sets at Westport, CT, were statistically different, as shown in Table 6. Essentially, a high F-value, and thus a correspondingly low p-value, indicates that the null hypothesis of both means being equal (H_o: $\mu_{STN}=\mu_{SOAP}$) must be rejected. The measurements at Queens, and the OC measurements at Westport, CT, however, showed that for those samples, the SOAP and STN mass concentrations were in statistical agreement.

Table 6- Results of F-tests Comparing SOAP and STN Ambient Concentrations per Monitoring Site

Chester		**F-Value**	**p (alpha=0.025)**
	OC	6.71	0.0107
	EC	19.14	2.40E-05
Elizabeth			
	OC	11.24	0.001
	EC	9.74	0.0022
Queens			
	OC	1.33	0.251
	EC	3.42	0.066
Westport			
	OC	0.78	0.3787
	EC	30.23	0

This statistical data is consistent with the results of the blank OC and EC mass levels where the STN blanks were estimated to be a higher percentage of the reported ambient concentrations than the SOAP study. The F-test results also indicates the SOAP and STN OC and EC reported data sets are statistically different, with the positive blank values indicating a positive systematic bias with the STN filters.

Table 7- Linear Least Squares Regression Results for Inter-method Comparison

Chester, NJ

STN OC vs SOAP OC

	Value	95% Confidence Interval
β_1=	0.6825	(0.3507, 1.014)
β_0=	1.592	(0.6505, 2.534)
R^2=	0.2326	

STN EC vs SOAP EC

	Value	95% Confidence Interval
β_1=	0.1405	(-0.0007171, 0.2817)
β_0=	0.3867	(0.3276, 0.4457)
R^2=	0.06624	

Elizabeth, NJ

STN OC vs SOAP OC

	Value	95% Confidence Interval
β_1=	0.8356	(0.6277, 1.044)
β_0=	2.08	(1.024, 3.137)
R^2=	0.5143	

STN EC vs SOAP EC

	Value	95% Confidence Interval
β_1=	1.492	(0.9946, 1.988)
β_0=	3.317	(2.309, 4.325)
R^2=	0.7127	

Queens, NY

STN OC vs SOAP OC

	Value	95% Confidence Interval
β_1=	0.6048	(0.4215, 0.7881)
β_0=	1.112	(0.3961, 1.829)
R^2=	0.4044	

STN EC vs SOAP EC

	Value	95% Confidence Interval
β_1=	0.6655	(0.4883, 0.8428)
β_0=	0.1306	(-0.02007, 0.2813)
R^2=	0.4679	

Westport, CT

STN OC vs SOAP OC

	Value	95% Confidence Interval
β_1=	0.8881	(0.6686, 1.108)
β_0=	0.7356	(0.01125, 1.46)
R^2=	0.6449	

STN EC vs SOAP EC

	Value	95% Confidence Interval
β_1=	1.321	(0.9868, 1.655)
β_0=	0.2199	(0.06457, 0.3752)
R^2=	0.6344	

B. Regressional Analysis:

In order to further compare the SOAP and STN reported ambient concentration data, linear least squares regression was performed in order to fit a scatter-plot of the two sample sets (Table 7). The curve-fitting toolbox from the mathematical software, Mathworks Matlab 7.4 was used for calculation of the regressions. The SOAP and STN data were fitted to a least squares regression line of the form as in the analysis of blank concentrations above:

$$Y_i = \beta_0 + \beta_1 X_i + \varepsilon_i$$

where X_i corresponds to the SOAP mass concentration and Y_i corresponds to the STN mass concentration. According to regression analysis (Kutner 2004), the null hypothesis would be that, since both the Tisch Sampler and the Met-One SASS sampler should give the same measured values in a 1:1 correspondence, the slope of the regression line would be unity (H_o: β_1=1).

Traditionally, in regression, the values of β_1 and β_0 are approximated by the estimator values of b_1 and b_0, respectively, though, in this context they will be used interchangeably. Those are calculated according to the formulae:

$$b_1 = \frac{\sum (X_i - \overline{X})(Y_i - \overline{Y})}{\sum (X_i - \overline{X})^2}$$

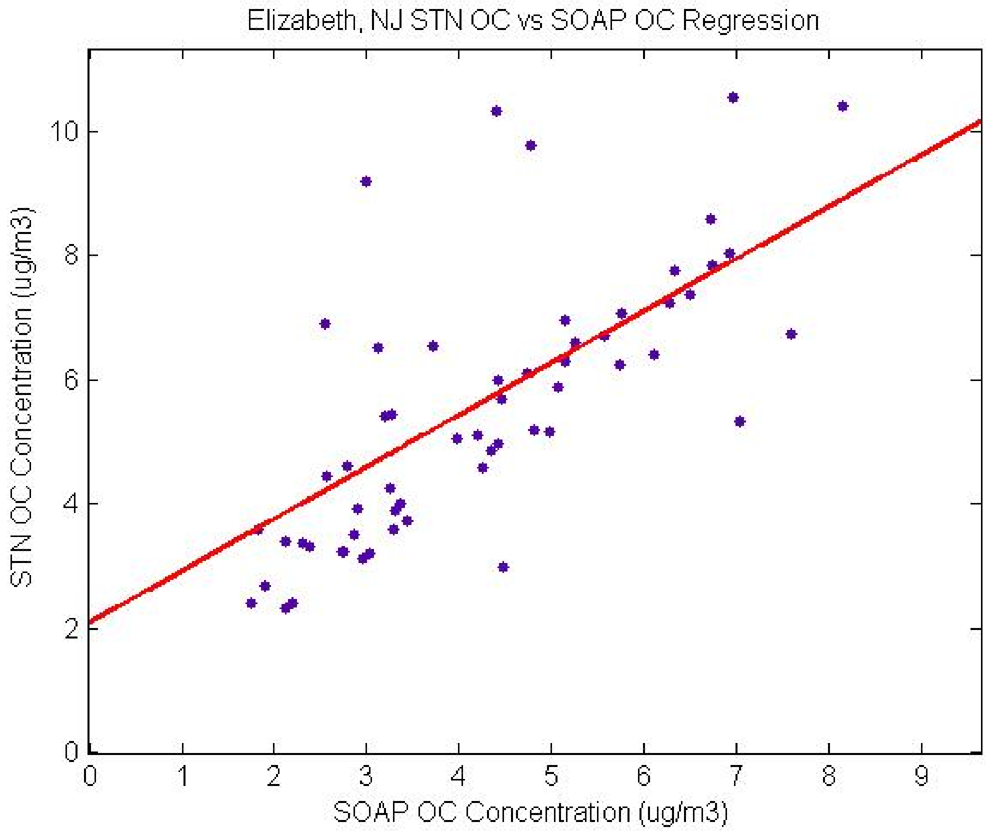
$$b_0 = \frac{1}{n}(\sum Y_i - b_1 \sum X_i) = \overline{Y} - b_1 \overline{X}$$

where X_i is a data point of the independent variable, and Y_i the matched data point of the dependent variable. The over bars indicate their mean values. In using approximated estimator values, it is assumed that actual slope and intercept of the fitted line lies in a 95% confidence interval about the approximated value. These are given by the equations:

$$\beta_1 \text{ lies within } b_1 \pm t(1-\alpha/2); n-2)s\{b_1\}$$
$$\beta_0 \text{ lies within } b_0 \pm t(1-\alpha/2); n-2)s\{b_0\}$$

where the statistical t-test is performed at the α significance level (α=0.05, corresponding to 95% confidence level), with n-2 degrees of freedom. The t-value is then multiplied by the $s\{b\}$ term which indicates standard deviation.

Figure 4- Sample Regression Plot Comparing Measured STN OC Concentrations with SOAP OC Concentrations at Elizabeth, NJ



In order to compare the sample sets through regressional analysis, only sampling data from dates in which both SOAP and STN had successfully collected filters were used. As seen from Table 4, the R^2 values for the regression lines, indicate the accuracy of the fit are not very high, with the highest value for Elizabeth EC measurements at 0.71. Due to atmospheric conditions, seasonal differences in aggregate primary and secondary $PM_{2.5}$ emissions, air mass compositions of OC and EC are highly variable. It is seen in Table 7 that the slopes of the lines at Chester, Elizabeth, and Westport include the null hypothesis value of 1 within the 95% confidence interval. However, the lines do not pass through the origin at the same confidence interval, indicating that the EC and OC corresponding to the SOAP theoretical $PM_{2.5}$ mass of 0 μg /m^3 does not correspond with the STN EC and OC masses when the theoretical $PM_{2.5}$ mass is 0 µg /m^3. At Queens, the slopes of the regression lines for OC and EC fitting do not include 1 in the 95% confidence interval. These statistical tests all point to the SOAP reported mass concentrations being statistically less than the STN reported measurements.

The SOAP and STN studies both utilized the same carbon analysis thermal optical transmission (TOT) method, NIOSH 5040. The consistently low SOAP OC mass concentrations would not be attributed to under measurement by the NIOSH 5040 method, since the analysis procedure is well monitored by a variety of blank analyses and calibrations using standards. One likely factor that would account for the difference in OC ambient mass concentrations between the SOAP and STN filters is the use of different samplers with different flow rates and filter exposed areas. These differences in samplers would effectively produce different face velocities where the more volatile OC species would be removed from the $PM_{2.5}$ mass collected on the filter surface (negative artifact effect). The Tisch sampler in the SOAP study operated at 113 liters/minute, corresponding to a daily intake of 162.72 m^3 of air, while the STN Met-One SASS sampler operated at a flow 16.7 L/min, with a total air intake of 24 m^3. Using the given areas of the circular quartz filters the total face velocity can be calculated Table 8). Turpin et al. 2000 found that an increased face velocity corresponded to lower measurements of OC, where a face velocity of 40 cm/s resulted in 22% less OC measured than a face velocity of 20cm/s.

Table 8- Calculation of SOAP, STN Face Velocities

Network	Sampler	Flow Rate (L/min)	Q Flow Rate (m^3/s)	A Filter Effective Area (m^2)	Face Velocity (m/s) (Q/A)	Face Velocity (cm/s)
SOAP	Tisch	113.00	0.0019	0.0057	0.3320	33.2000
STN	Met-One SASS	16.70	0.0003	0.0011	0.2463	24.6300

In Table 8 the Tisch sampler face velocity was determined to be approximately 33.26 cm/s using the calculation:

Face Velocity = [Flow Rate]/[Cross Sectional Area].

The Met-One SASS sampler face velocity was calculated as 24.63 cm/s, approximately 25% less. Therefore, at least part of the lower SOAP OC mass concentrations are due to differences in sample collection.

III. Trends

With reference once more to Table 5 the measures of central tendency depict spatial and temporal trends in EC and OC ambient mass concentration. Gray et al. 1998 showed that it is possible to attribute urban EC or black carbon to motor vehicle traffic and fuel combustion (home fireplaces, wildfires, structural fires, industrial fires). The EC levels for the four sites steadily increase from less trafficked, less industrial sites to the highest. Chester, NJ exhibits the lowest elemental carbon concentration, followed by Westport, CT, then Queens, NY, and finally Elizabeth, NJ with the highest concentration of elemental carbon on average. The Elizabeth, NJ, monitoring site at the NJ Turnpike toll plaza has 5 to 7 times more elemental carbon ambient mass (μg/m3), than the Chester

New Jersey site. Ratios of Elizabeth EC to the means and medians for other sites are given in Table 9.

Table 9- Ratios of the SOAP Average Ambient EC Concentrations To Elizabeth Means and Medians

	Means	Medians
Chester	5.74	6.51
Queens	2.03	2.07
Westport	3.31	3.39

Table 10- Ratios of the STN Average Ambient EC Concentrations To Elizabeth Means and Medians

	Means	Medians
Chester	1.77	2.05
Queens	1.31	1.39
Westport	1.46	1.50

The difference in the OC and EC levels at Elizabeth compared to the other sites may simply reflect the overwhelming proportion of transportation emissions at this site compared to the contributions of other primary sources and secondary OC at the other sites. An evaluation of the molecular markers present as OC at Elizabeth and the three other SOAP sites will give great insight on the sources contributing to the $PM_{2.5}$ mass. The molecular marker composition of the OC mass component is reported in other papers emanating from the SOAP study. It should also be noted that in the US Northeast corridor there is a prevailing wind blowing across the Ohio Valley, through the New York Metropolitan Area, and beyond further Northeast (McDow et al. 2007). Thus, the Westport, CT site, which is downwind from the Queens site, which itself is downwind from the Chester site could possibly be the recipient of aged organics along the air mass trajectory (Qin et al. 2006). Chester, NJ receives regional aged aerosol from the Ohio Valley, Washington D.C., and Philadelphia, PA.

Ambient time series data comparing OC, EC and $PM_{2.5}$ mass for are plotted in Figures 5 through 6. The ambient plots for SOAP OC and EC are consistently lower than the STN concentrations. These differences can be attributed in part to the different face velocities of the samplers, with the exception of the Queens plots where the two times series virtually coincide. At each site and for each network, the daily time series plots exhibit temporal mass variations in OC, EC, and $PM_{2.5}$ mass, possibly due to differing air mass trajectories, seasonal emissions, and prevailing atmospheric conditions (height of the mixed layer). A much smoother plot of the OC and EC concentrations was made when seasonal averages were plotted (Figures 7 through 10). In these plots, the numerical averages were calculated by grouping the 24-hour daily concentration measurements according to the same composite scheme as the seasonal composites used for analytical molecular marker measurements specified in the SOAP sampling protocol (McDow et al 2007). This allowed for visualization of trends otherwise obscured by the time series noise.

Figure 5- Daily Ambient OC Concentrations (µg /m3)

(a) Chester, NJ (b) Elizabeth, NJ (c) Queens, NY (d) Westport, CT

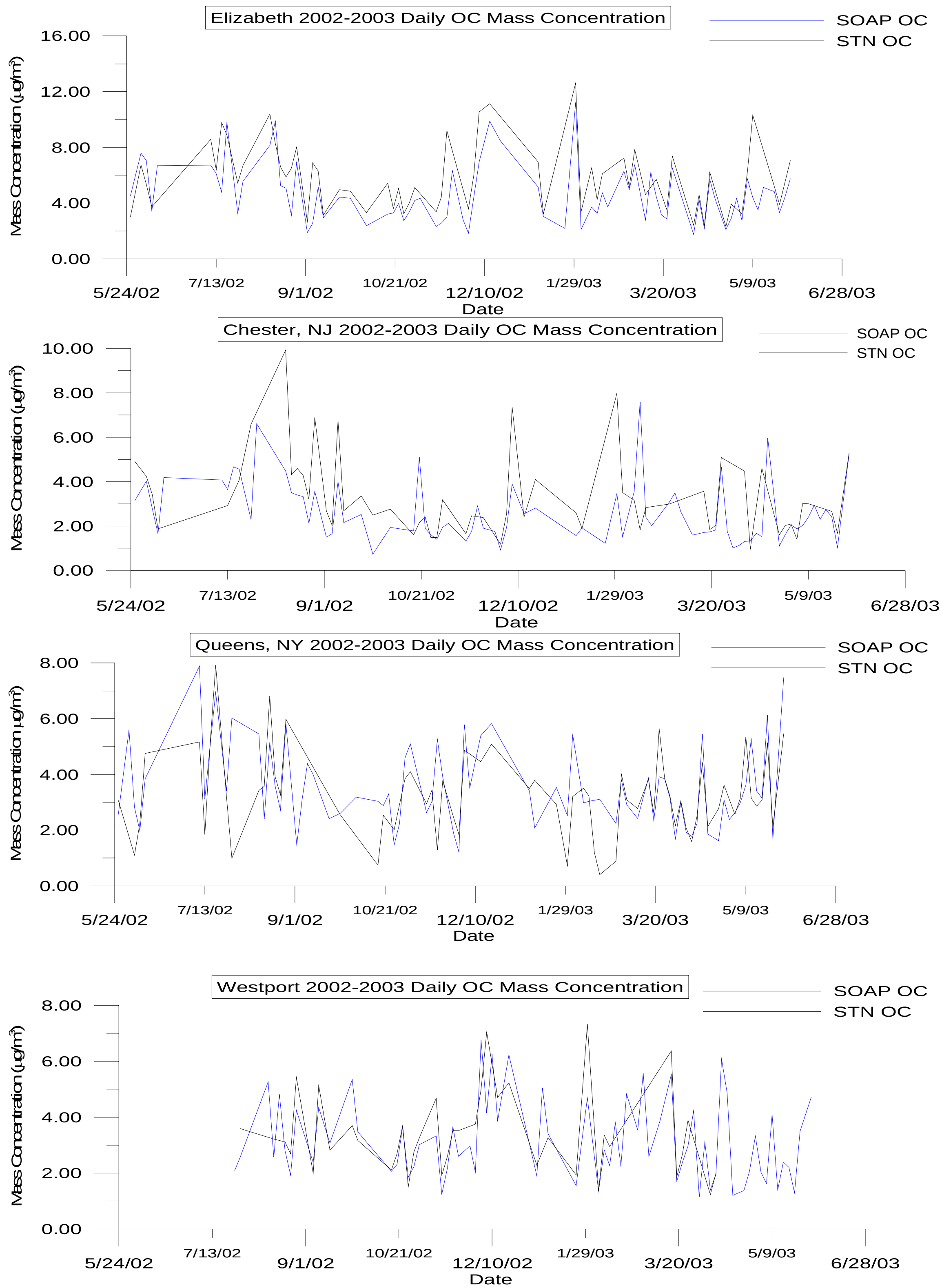


Figure 6- Daily Ambient EC Concentrations (µg /m3)

(a) Chester, NJ (b) Elizabeth, NJ (c) Queens, NY (d) Westport, CT

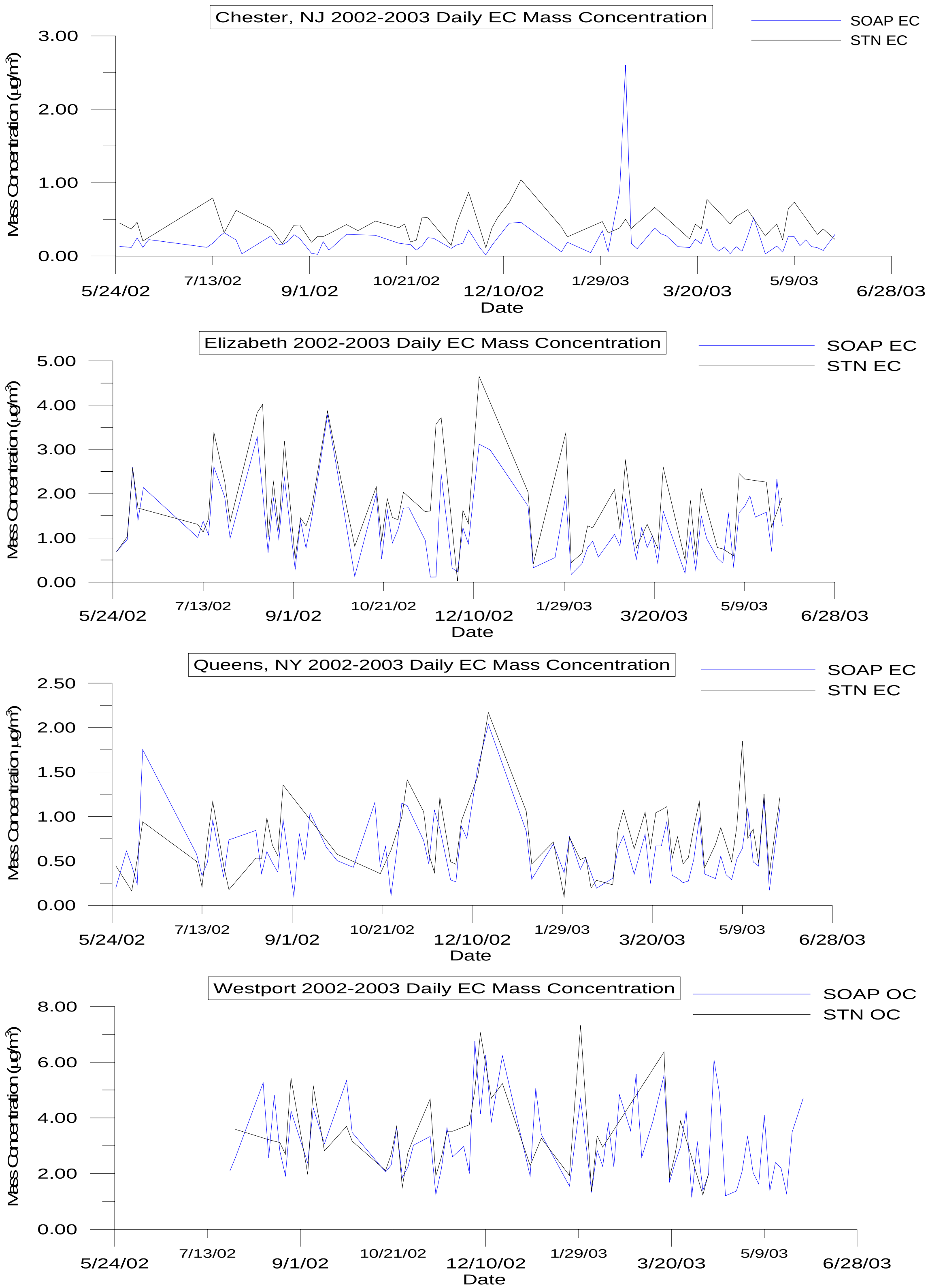
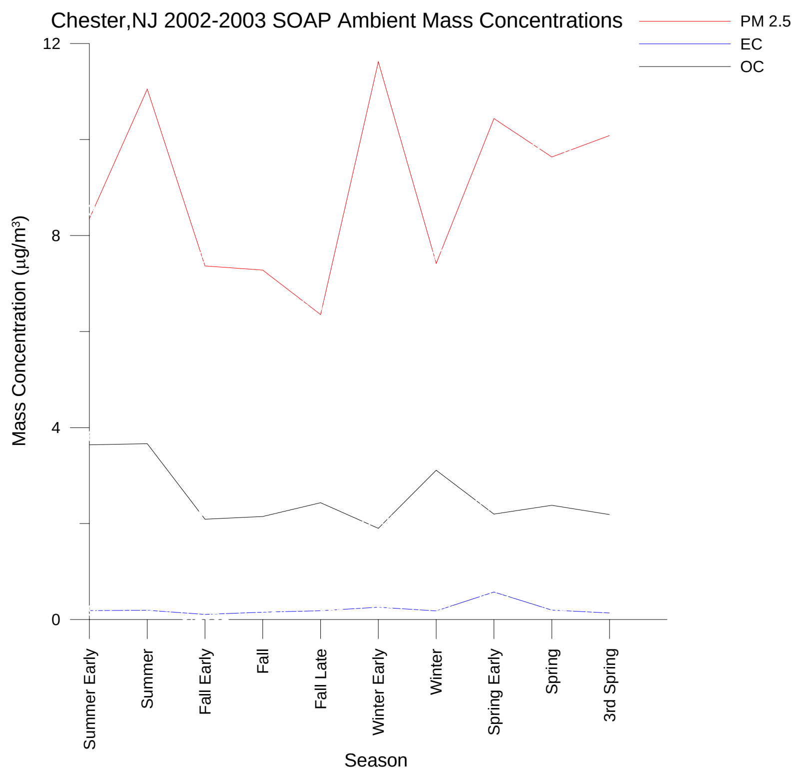


Figure 7- SOAP Seasonal Mass Concentrations

(a) Chester, NJ (b) Elizabeth, NJ



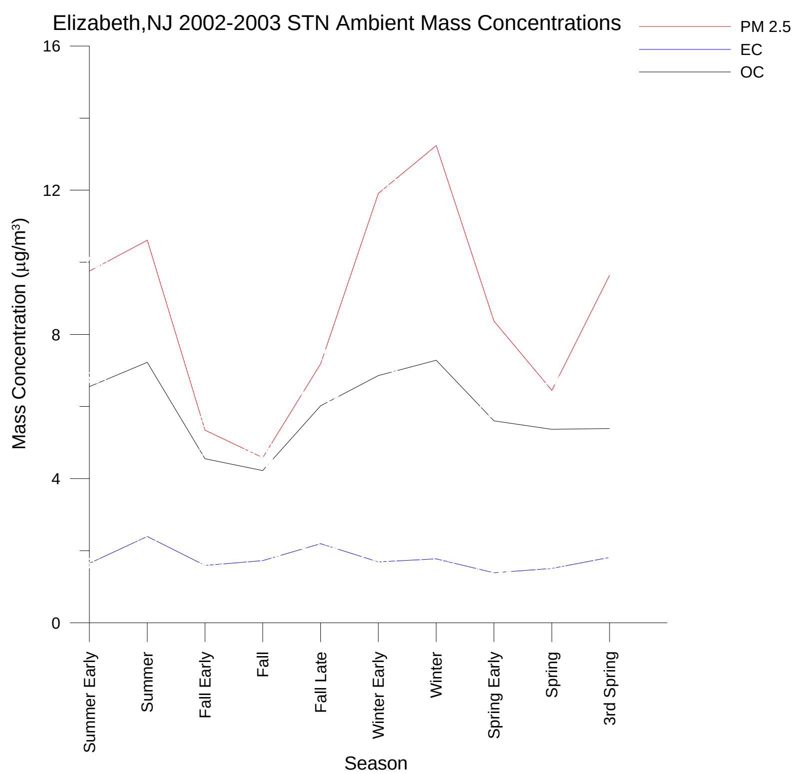
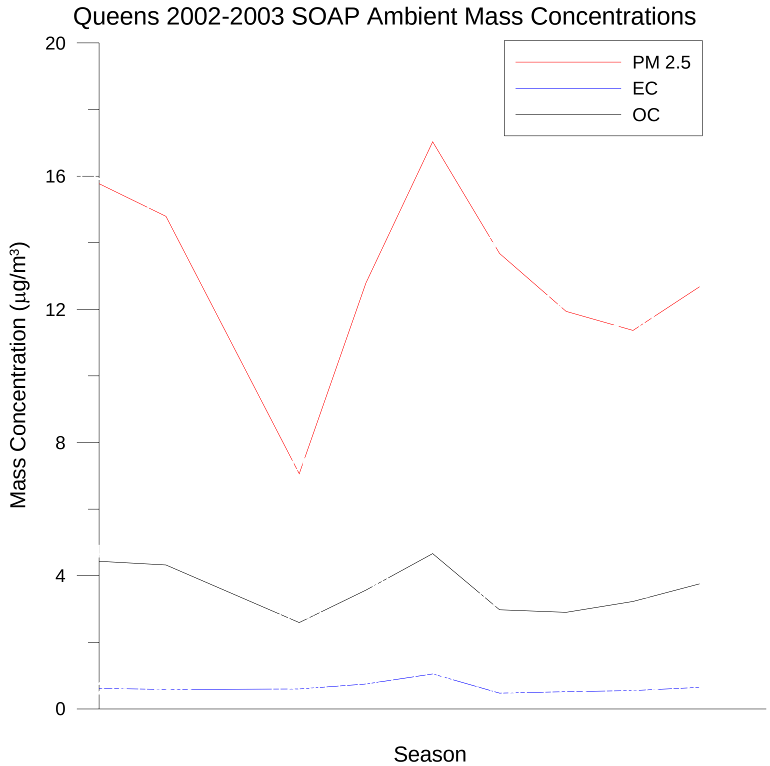


Figure 8- SOAP Seasonal Mass Concentrations

(a) Queens, NY (b) Westport, CT



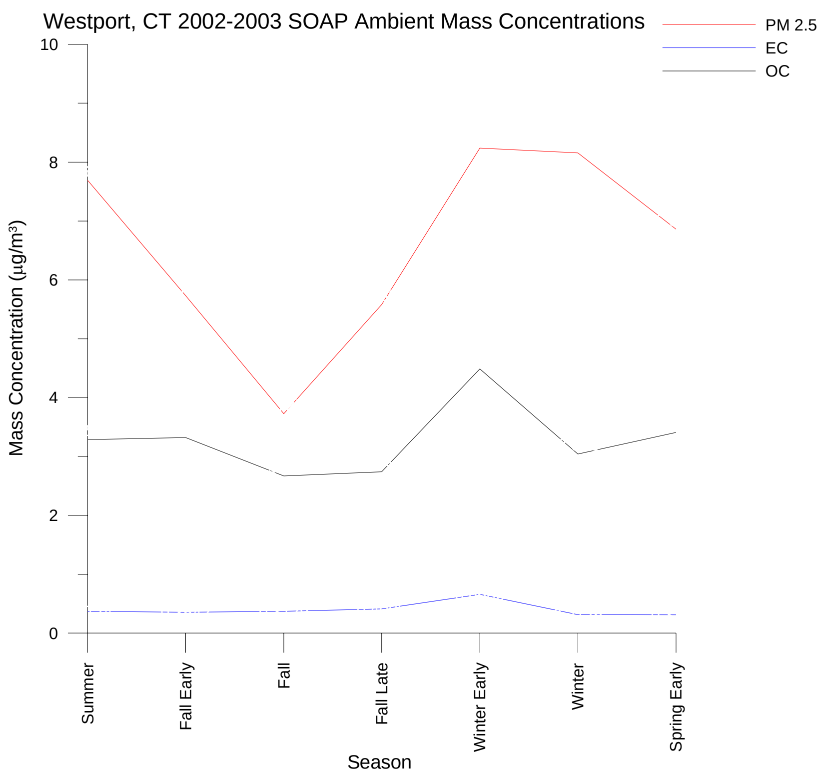
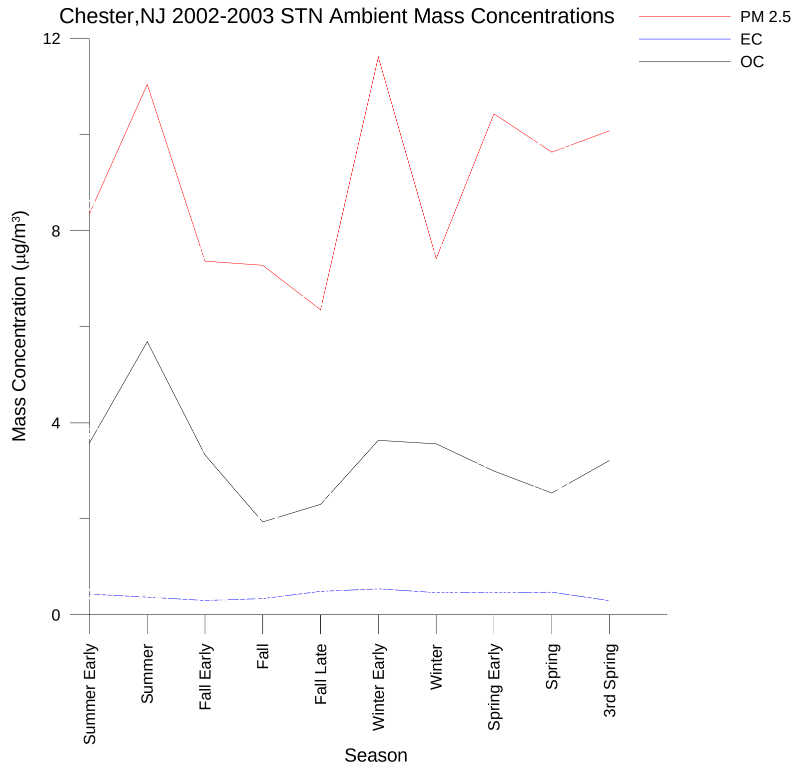


Figure 9- STN Seasonal Mass Concentrations

(a) Chester, NJ (b) Elizabeth, NJ



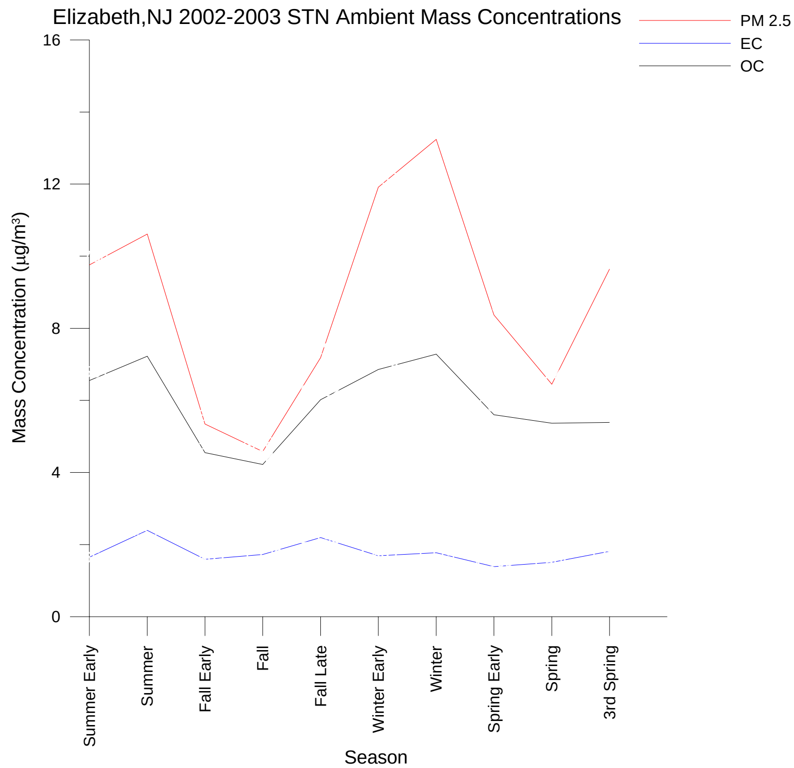
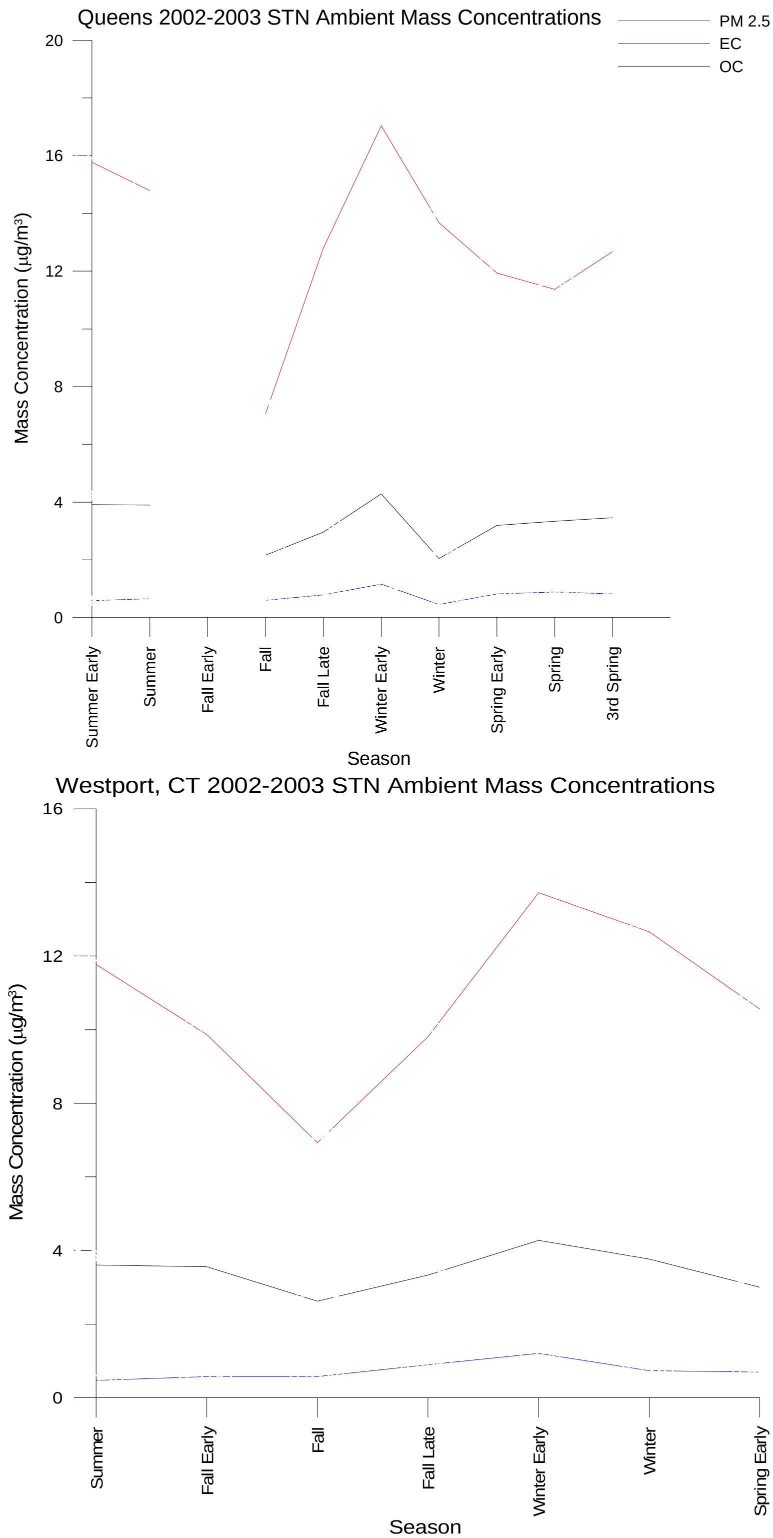


Figure 10- STN Seasonal Mass Concentrations

(a) Queens, NY (b) Westport, CT



In Figures 7-10 the trends for all four sites show local maxima of organic carbon and elemental carbon ambient mass concentrations during the winter seasons (early winter, winter, late winter). Each site also has an additional peak in organic carbon for the summer seasons (early summer, summer), whereas it appears only Elizabeth, NJ exhibited an additional EC peak in summer peak. It is significant to note the Chester, NJ was the only site in which the summer peak was higher than its winter peak. The EC and OC plots for Elizabeth, NJ show parallel trends throughout the annual cycle, compared to Chester which has the OC peak in the summer and the EC peak in the winter. The parallel tracking of OC and EC at Elizabeth points to a strong local input by vehicular traffic that is fairly consistent throughout the year. Conversely, divergent EC and OC annual trends at Chester suggest additional local sources of OC during the summer months, such as photochemical reactions, and in addition to long-range transport of aged aerosol from upwind sources. It can be noticed that both Chester and Westport also have OC peaks in the summer, possibly indicating a larger degree of secondary OC present at Westport as well.

Elemental carbon particles are contributed by primary combustion sources such as motor vehicles, manufacturing, and wood burning, whereas OC can also be formed in the atmosphere by photochemical reactions of manmade and biogenic volatile organic carbons. If the EC sources were due only to motor vehicle traffic, and the transportation sector, more generally, then a fairly uniform seasonal concentration of OC and EC mass would be seen over the course of the year and varying with the height of the mixed layer (NARSTO 2004). However, the increase in OC and EC during winter might also indicate the increased seasonal emissions of wood burning from home fireplaces in the sampling area and the upwind source areas. This would explain the winter peaking observed at all four monitoring sites. As The Elizabeth, NJ site is adjacent to an area of heavy traffic flow on the NJ Turnpike, however, peaks in the summer (between July 25th, 2002 and August 27th, 2002), could also coincide with increased summer highway travel.

Analysis of the differences of trends in EC and OC gives some insight into emissions of fine particles from non-mobile OC sources and from secondary organic formation. Supplementing the time series plots per site of the seasonal average is a series of box and whisker plots (Figures 11 through 18). The box plots, generated by the Matlab 7.4 statistical toolbox, depict the range between the 75th and 25th percentiles of ambient concentration values within the box, with the median indicated by the line within the box. The whiskers extend to the smallest values within 1.5 times the inter-quartile range (IQR, calculated as the numerical difference between the 75th and 25th percentile values), with outliers indicated by the (+) sign. These plots show the variability in the data for a given season about its median.

From the box plots of the seasonal trends, it can be seen that for the Chester site, the elemental carbon concentrations generally stay within a similar inter-quartile range over the course of the year. Chester exhibits a distinctly high IQR in organic carbon for the summer seasons, but less distinct IQRs according to both STN and SOAP protocols in the winter months. Elizabeth shows a similar IQR between the early summer/summer and the late fall/early winter/winter in both its EC and OC seasonal concentrations. Consistent with the close proximity to the NJ Turnpike, the OC peaks correspond to EC peaks when both means and medians are investigated. The strength of the relationship of OC and EC mass concentrations at Elizabeth were analyzed via linear least squares regression where the SOAP R^2=0.40 and the STN R^2=0.37. Statistically, only a fair correlation is present between the ambient OC and EC fine PM components.

The Queens monitoring site has an IQR for the EC daily concentrations that is relatively constant over the year. An elevated IQR for EC is present in the early winter. There is a corresponding peak in OC in early winter, according to the box and whisker plots. The plots also show the IQR in early summer and summer is roughly parallel in OC to that of the winter peak. As the EC concentrations in the summer at Queens do not seem elevated relative to the rest of the year, the increase in OC median and variability in the warm months could be attributed to a higher proportion of secondary OC mass. The box and whisker plots show parallel winter median peaks for both SOAP and STN EC and OC concentrations. The increase in OC and EC during winter at the Queens site could indicate seasonal differences in primary emission sources where would smoke contributes a higher proportion to fine particle mass. Westport also has the highest median OC and EC ambient concentrations in winter, which also could be attributed to increased wood burning in the NY City area as well as in the upwind sites.

Figure 11- Chester, NJ SOAP 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC

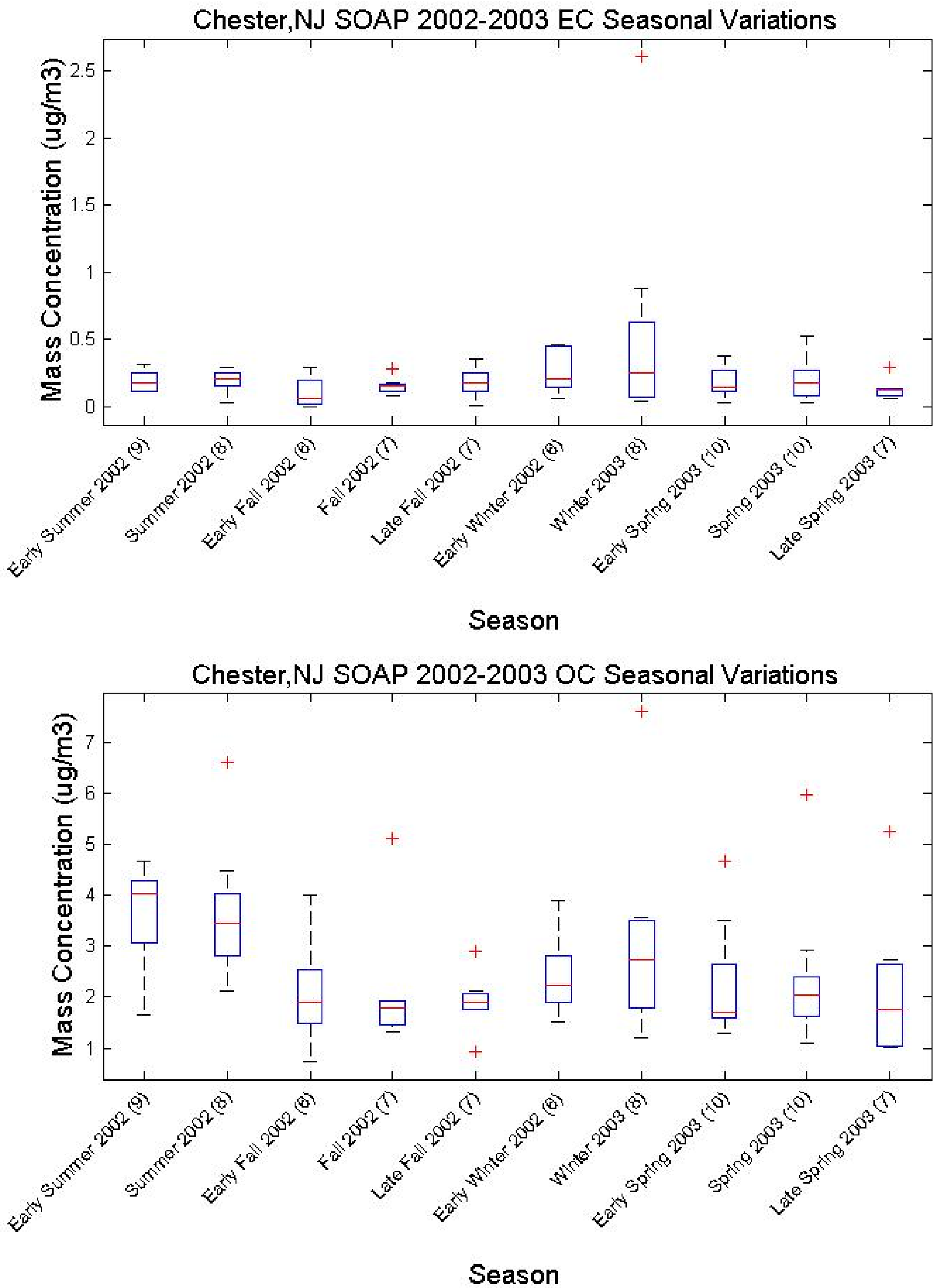
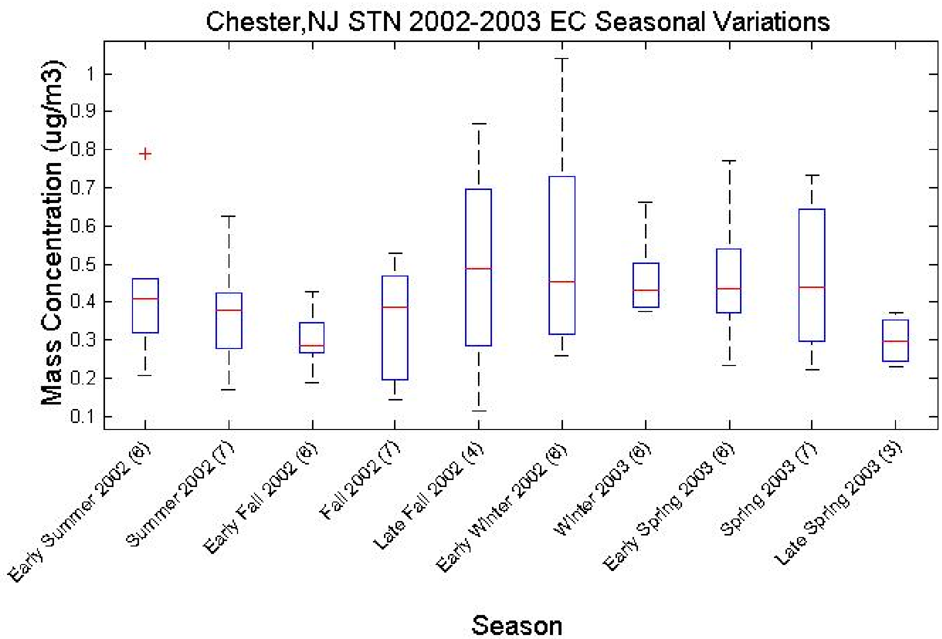


Figure 12- Chester, NJ STN 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC



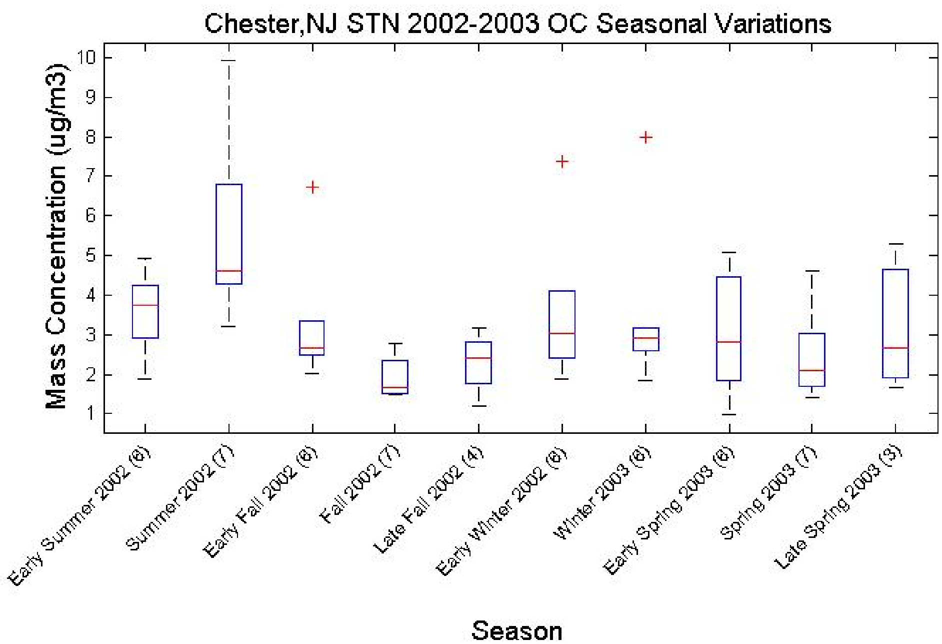


Figure 13- Elizabeth, NJ SOAP 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC

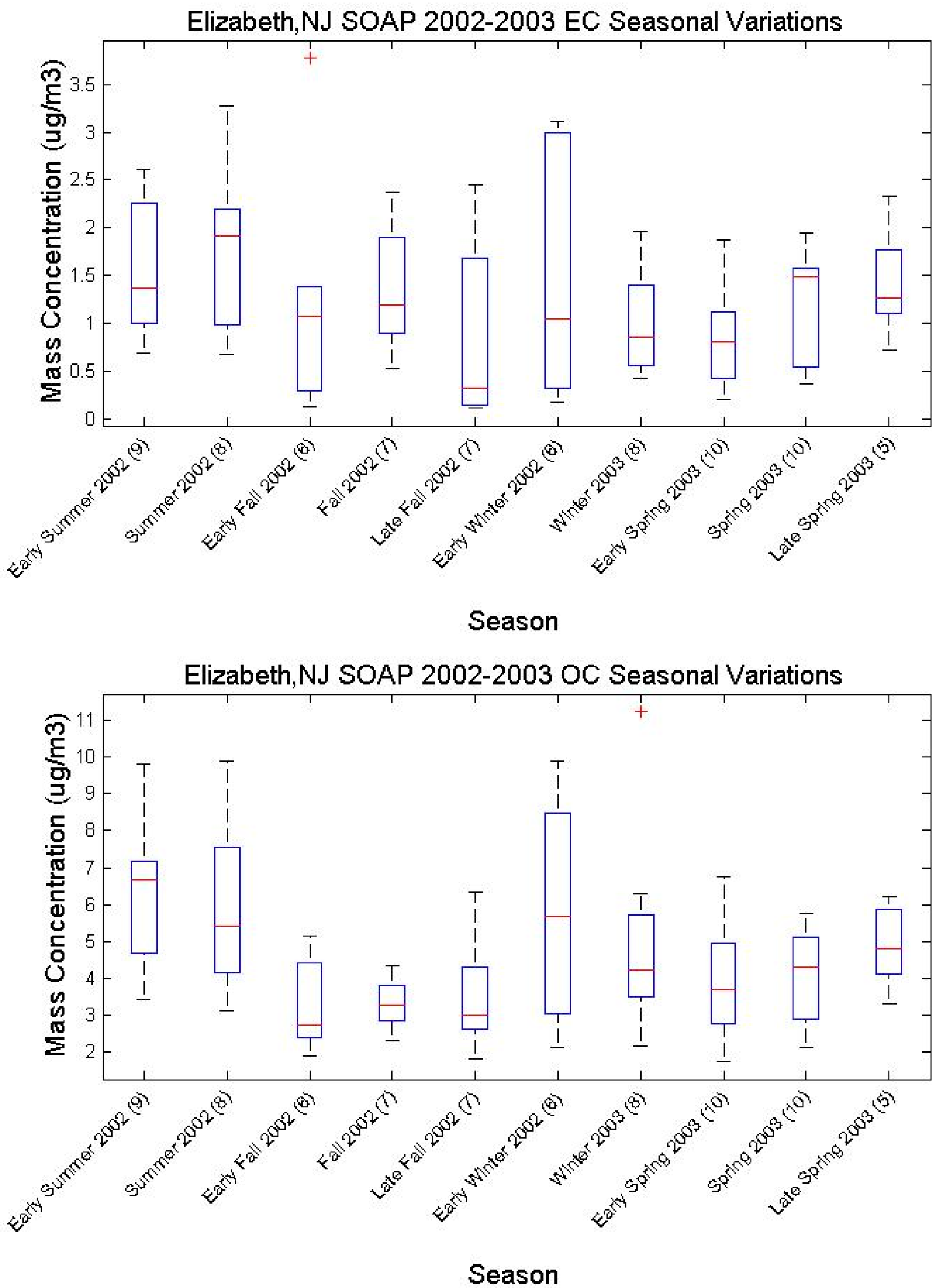


Figure 14-Elizabeth, NJ STN 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC

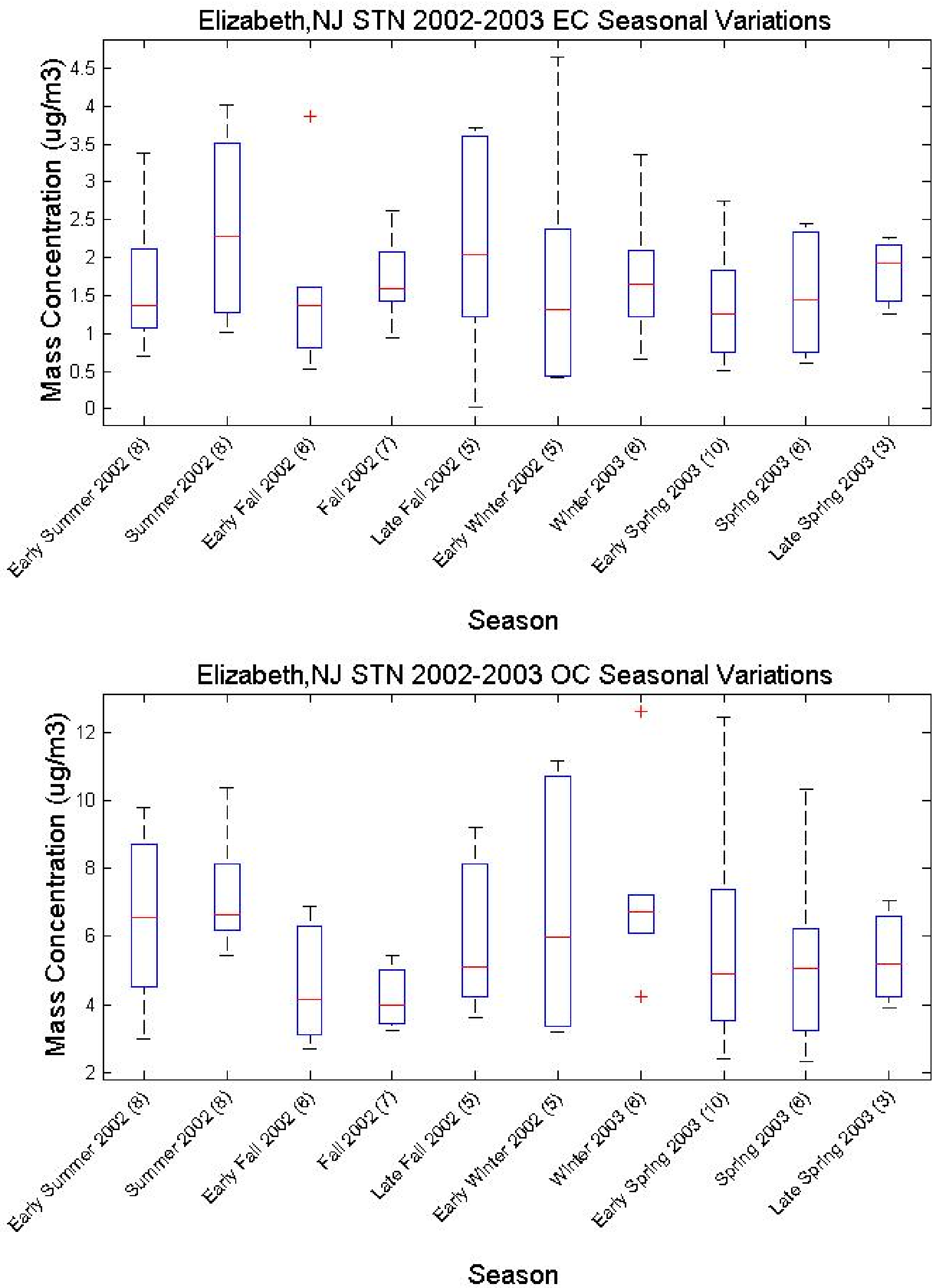
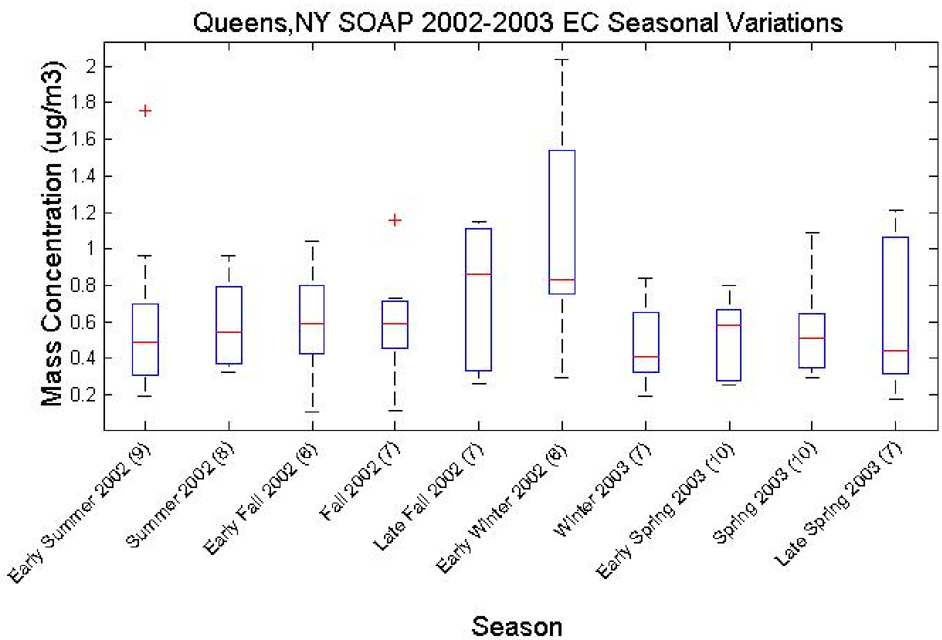


Figure 15- Queens, NY SOAP 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC



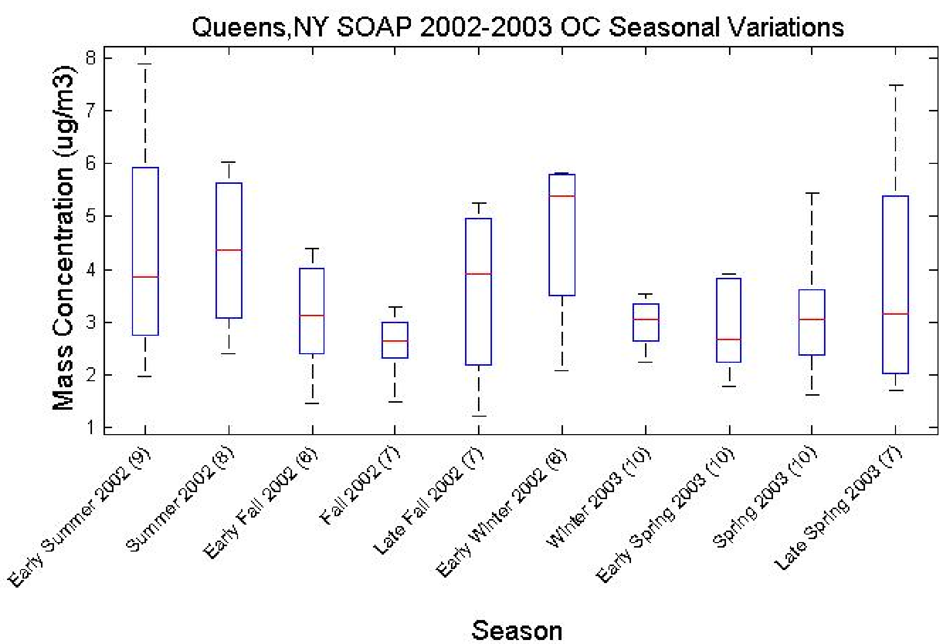
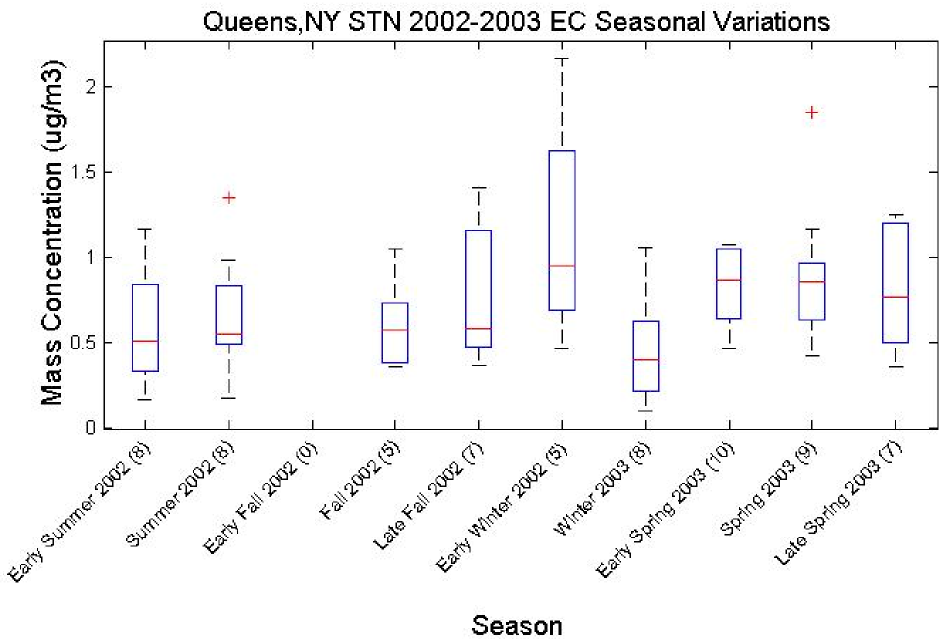


Figure 16- Queens, NY STN 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC



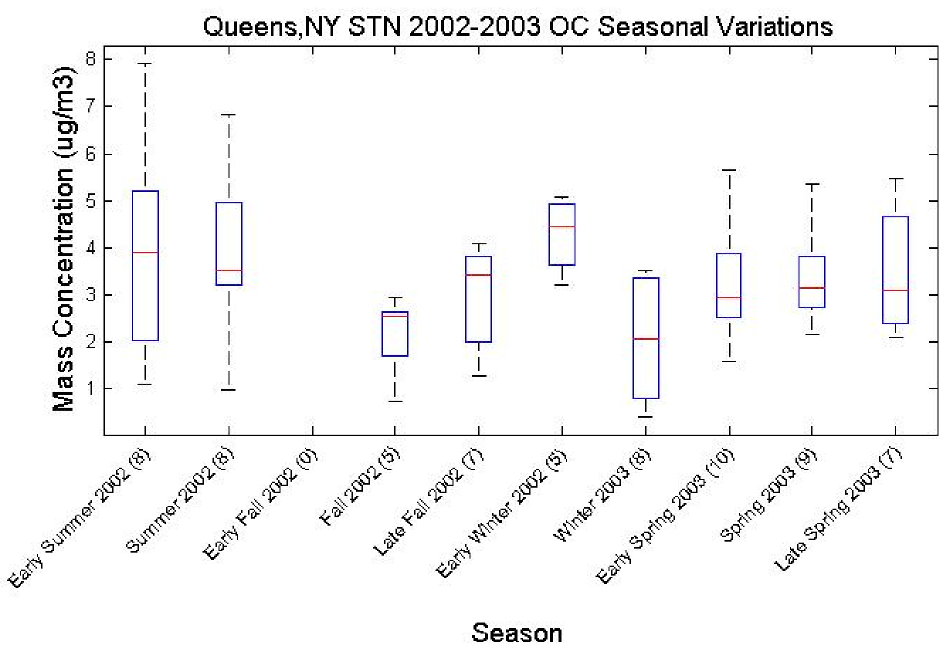
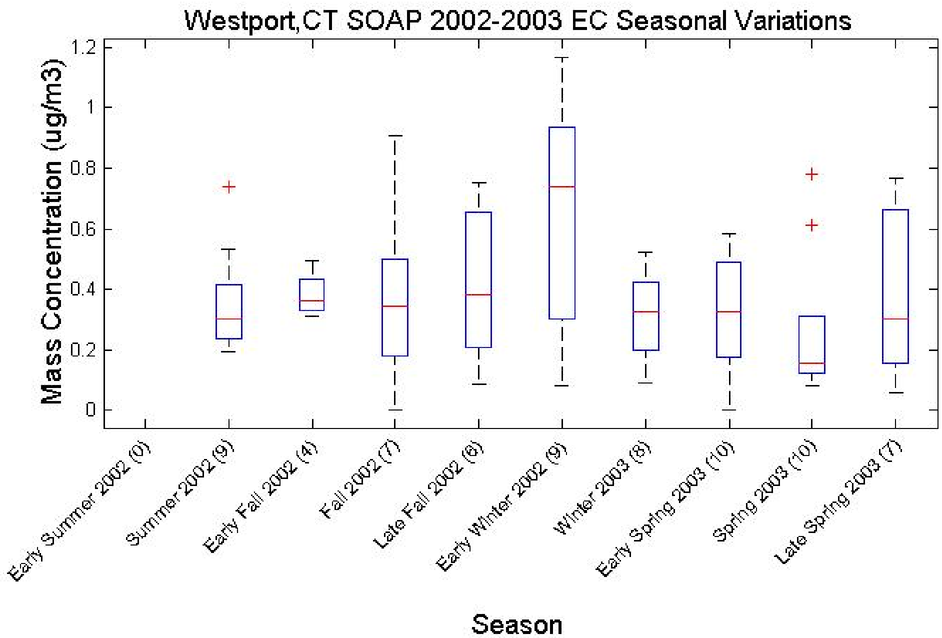


Figure 17- Westport, CT SOAP 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC



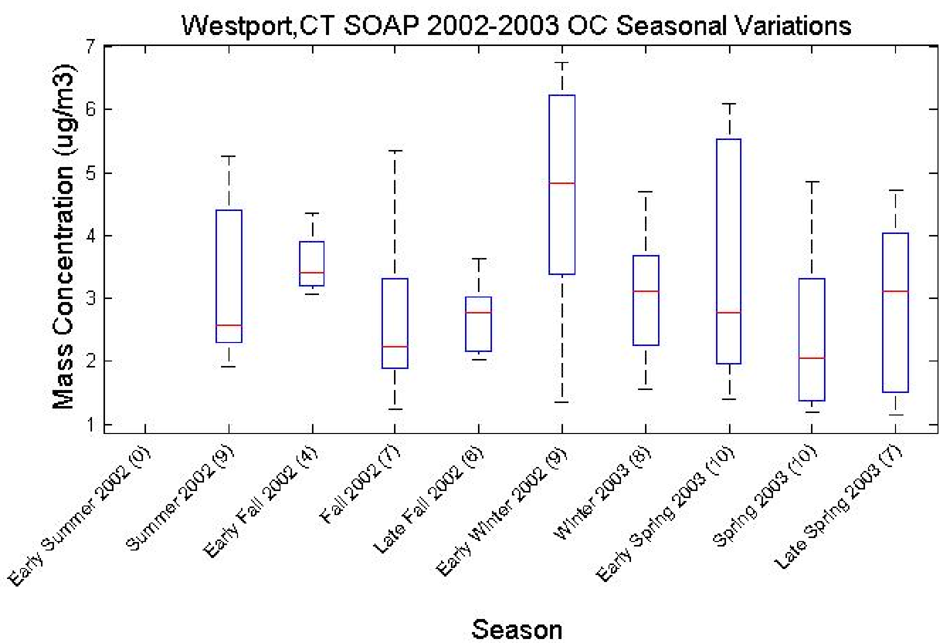
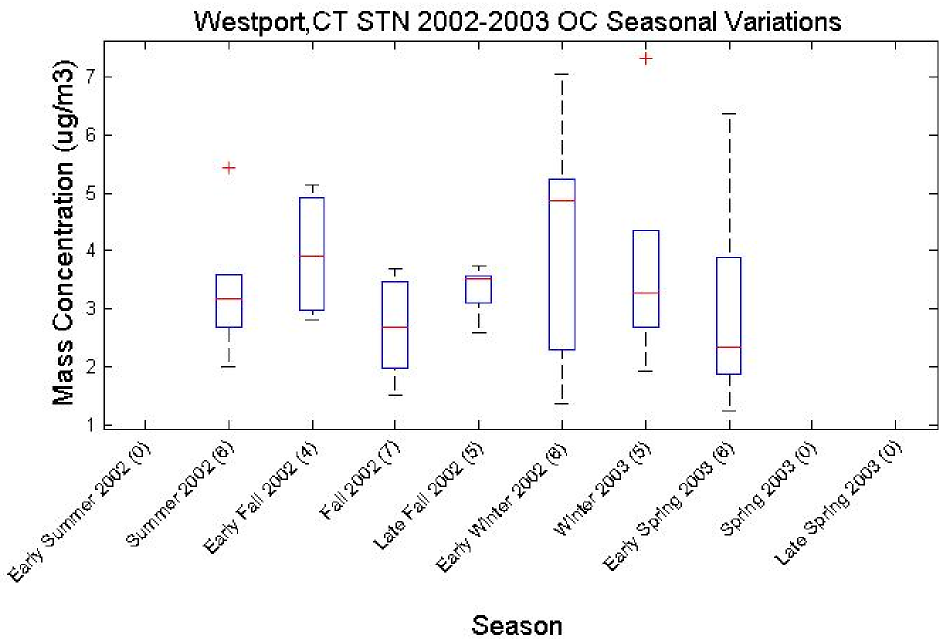
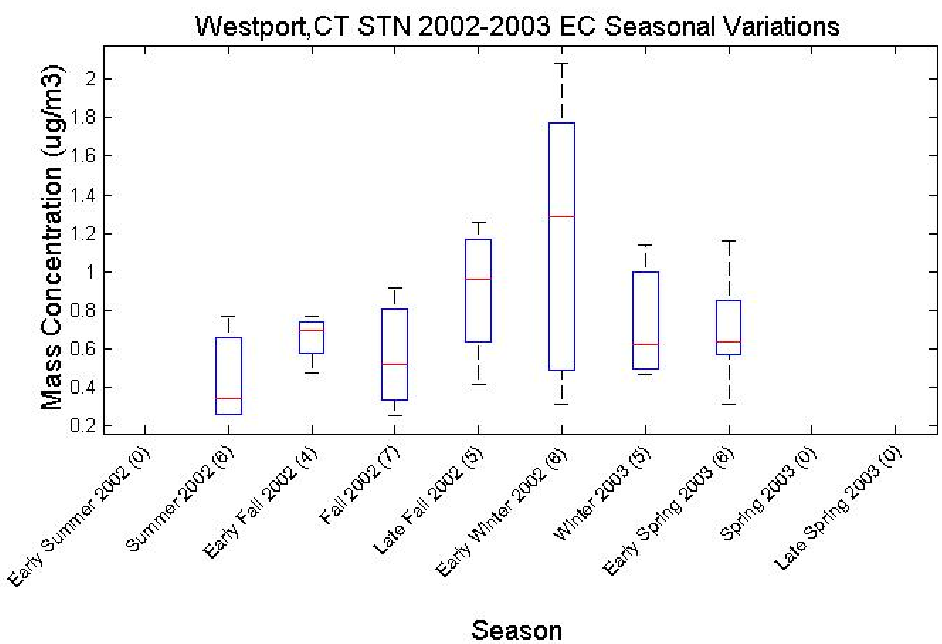


Figure 18- Westport, CT STN 2002-2003 Box and Whisker Plots Indicating Inter-Quartile Range and Outlier Values of Ambient Concentrations for Seasonal (a) EC and (b) OC





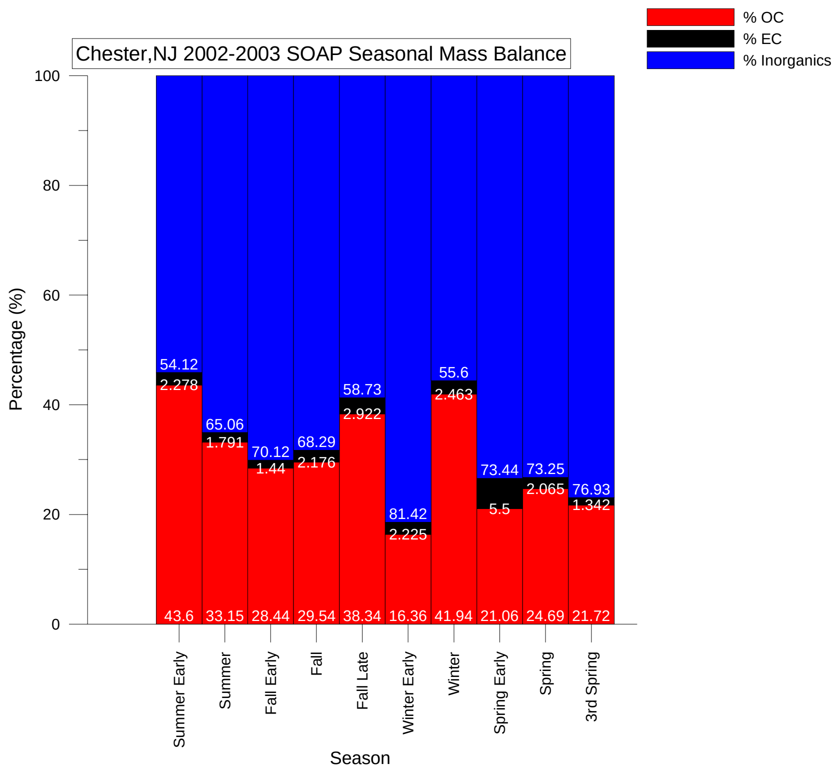
IV. Seasonal Mass Balances

Figures 19 through 22 show the chemical mass balances for the SOAP and STN fine particles in terms of OC, EC, and inorganic (and other) components. The mass balances were calculated using the STN $PM_{2.5}$ mass concentrations, averaging those over the seasons used in the SOAP composites, and subtracting average OC and EC concentrations to determine the non-carbonaceous portion of $PM_{2.5}$. The calculation is given in the expression below:

$$[\text{Inorganic + Others Avg.}] = [PM_{2.5}\ \text{Conc.}] - \{[\text{Elemental Carbon Conc.}] + [\text{Organic Carbon Conc.}]\}$$

where the brackets [] denote concentrations. The averaged value of each species was divided by the averaged total $PM_{2.5}$ to determine the percentage composition per season. This simple analysis was performed for the SOAP and STN seasonal composites.

Figure 19-Chester, NJ 2002-2003 Seasonal Mass Balance for (a) SOAP (b) STN



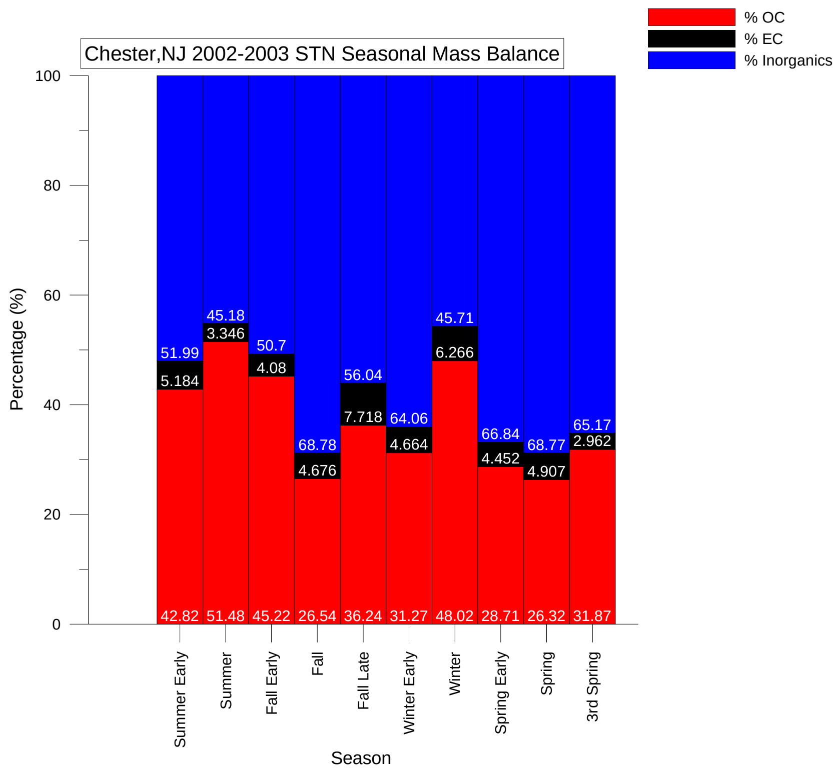
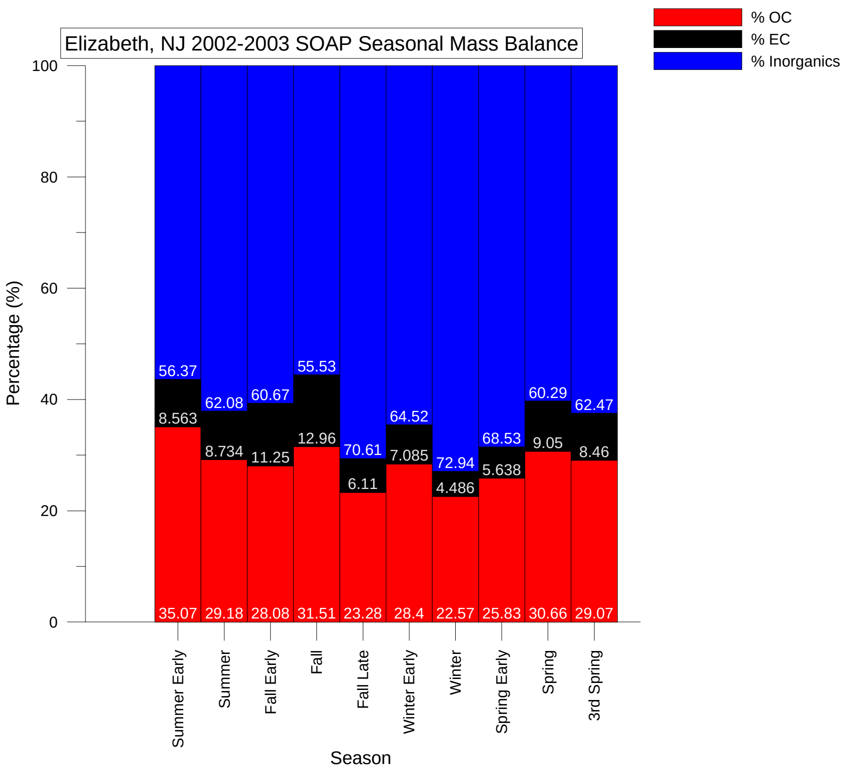


Figure 20- Elizabeth, NJ 2002-2003 Seasonal Mass Balance for (a) SOAP (b) STN



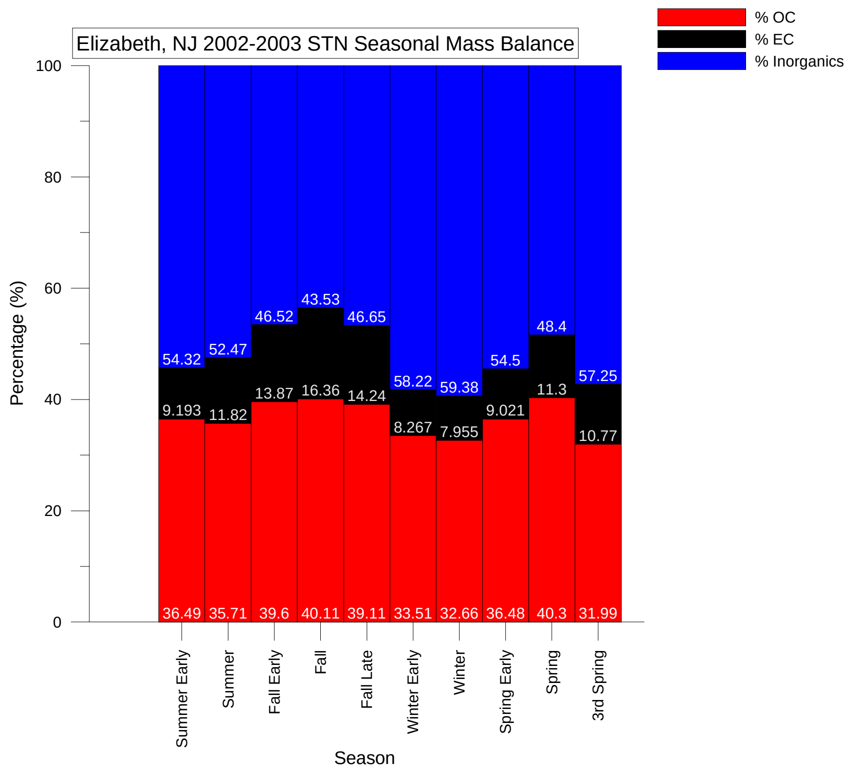


Figure 21- Queens, NY 2002-2003 Seasonal Mass Balance for (a) SOAP (b) STN

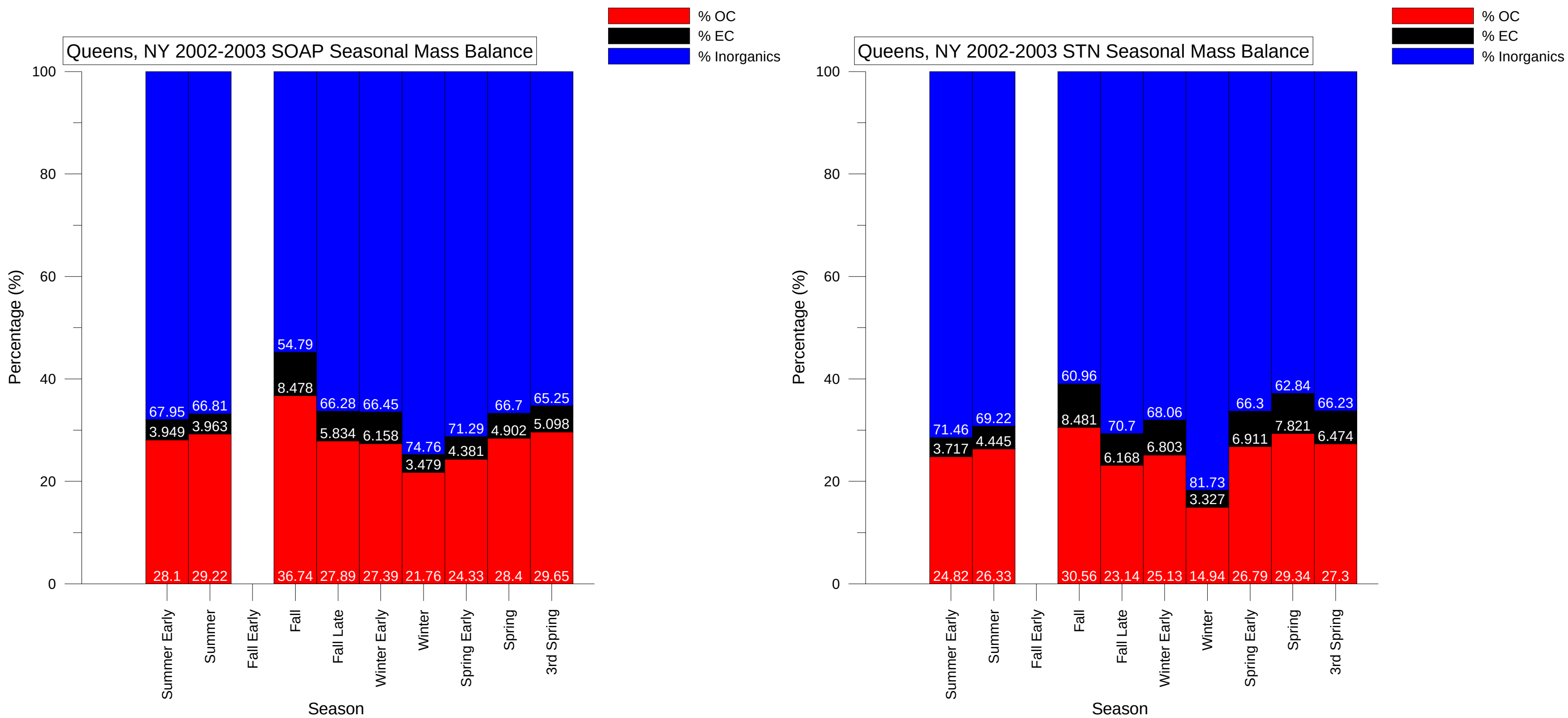
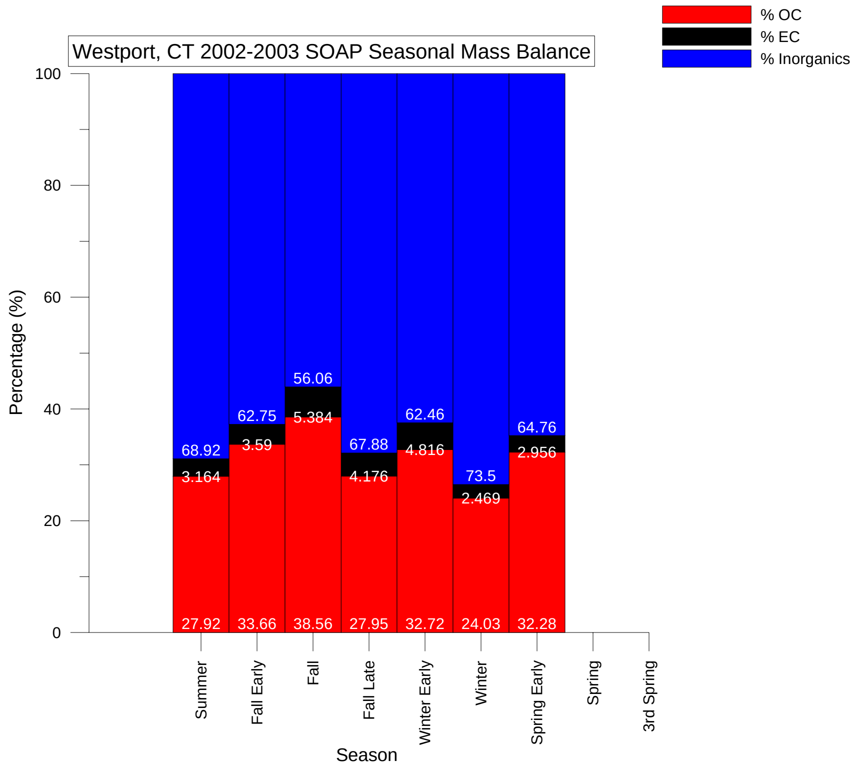
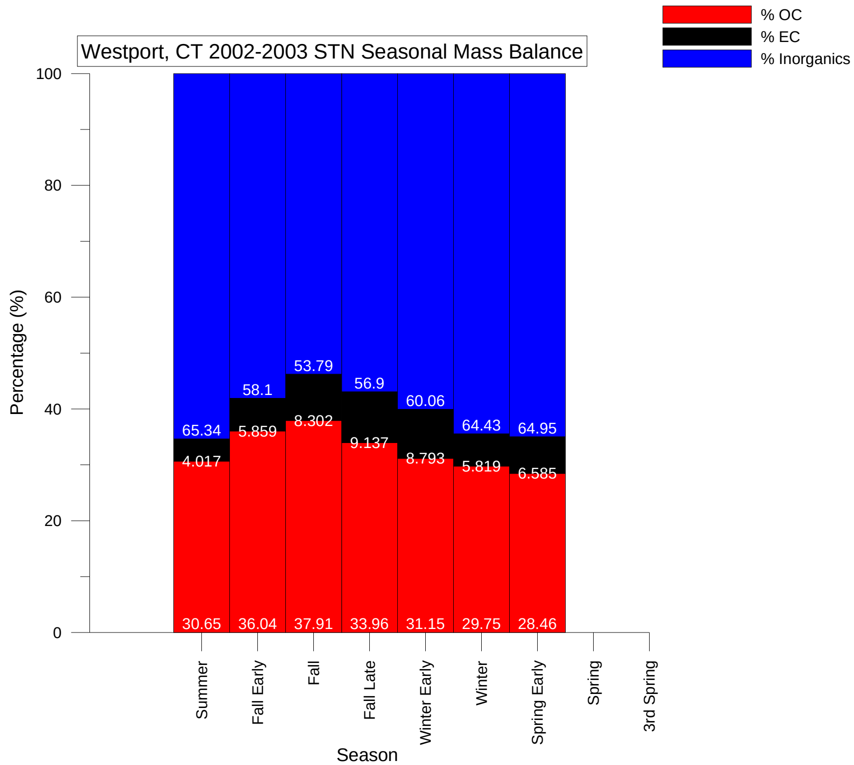


Figure 22- Westport, CT 2002-2003 Seasonal Mass Balance for (a) SOAP (b) STN





The mass balance plots show the percentage composition of OC at Chester during both the summer and winter months approaches 50% of the total composition of $PM_{2.5}$. There is roughly a 30% difference between the summer peak and the fall minimum concentrations. The mass balance is much different when compared to the Elizabeth site, which shows the composition of OC being roughly 35% of the total $PM_{2.5}$ mass and is relatively constant over the entire year. The composition of OC and EC at Elizabeth appear to track well. The chemical mass balances for EC and OC at the Queens and Westport sites also demonstrate parallel patterns with slight increase in the fall months. Whereas the Chester OC, as a percentage of total $PM_{2.5}$ mass, appears to be independent of the EC component, the rest of the sites show a consistent proportion in the amount of EC and OC present.

V. OC/EC Ratios

In order to determine the proportion of primary to secondary OC present in the $PM_{2.5}$ mass, the trends in the ratio of OC to EC ambient mass concentrations must be investigated. As discussed earlier, EC can be used as a conservative tracer of primary emissions. Variations in atmospheric concentrations normalized to EC can be compared by constructing OC/EC ratios. Box and whisker plots were generated for the OC/EC ratios of the ambient time series concentrations above (Figures 23 through 26). For Chester the later spring through early fall show greater variability about the OC/EC ratio median, on an order of 2 to 5 times that of the OC/EC ratios at the other monitoring sites. Moreover, aside from the IQR being wider in the late fall, the OC/EC ratio variability at Chester remains fairly constant over the annual cycle. Queens, NY also has a peak during the early summer and summer, but it is about half the OC/EC ratio observed at Chester. The STN data indicate an elevated OC/EC ratio for the summer at Westport; however the SOAP data do not show as distinctive a peak. The rest of the STN data is incomplete for the other warm months for clear trends to emerge.

Figure 23- Chester, NJ Box-Whisker Plot of OC/EC Ratios based on Ambient Concentrations from (a) SOAP and (b) STN Sampling Studies

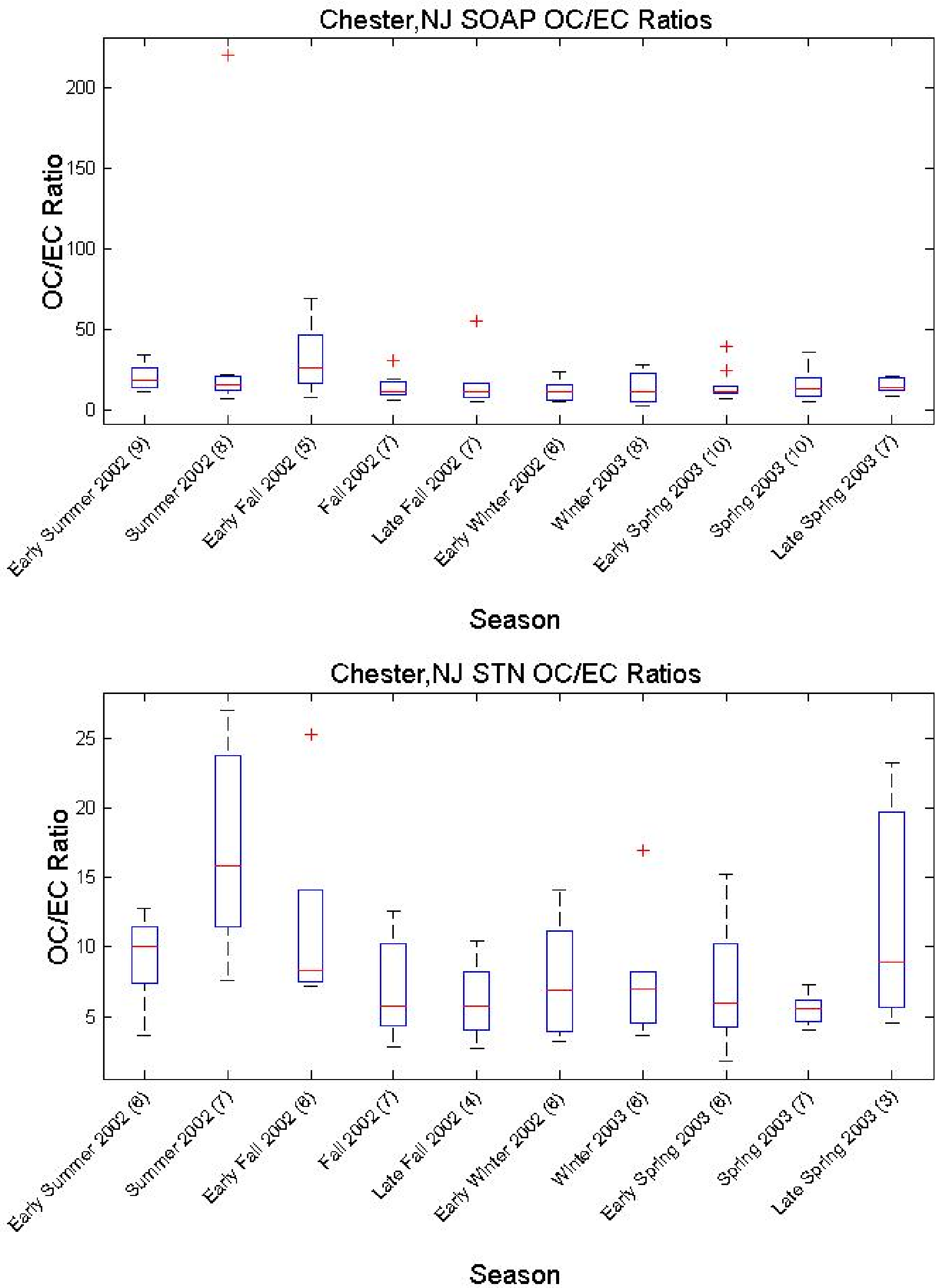


Figure 24- Elizabeth, NJ Box-Whisker Plot of OC/EC Ratios based on Ambient Concentrations from (a) SOAP and (b) STN Sampling Studies

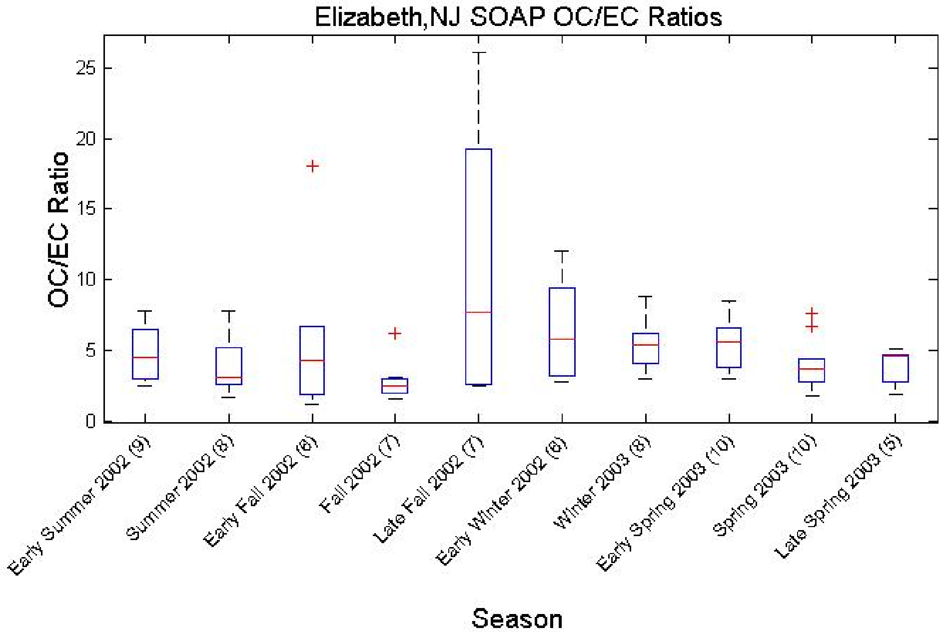


Figure 25- Queens, NY Box-Whisker Plot of OC/EC Ratios based on Ambient Concentrations from (a) SOAP and (b) STN Sampling Studies

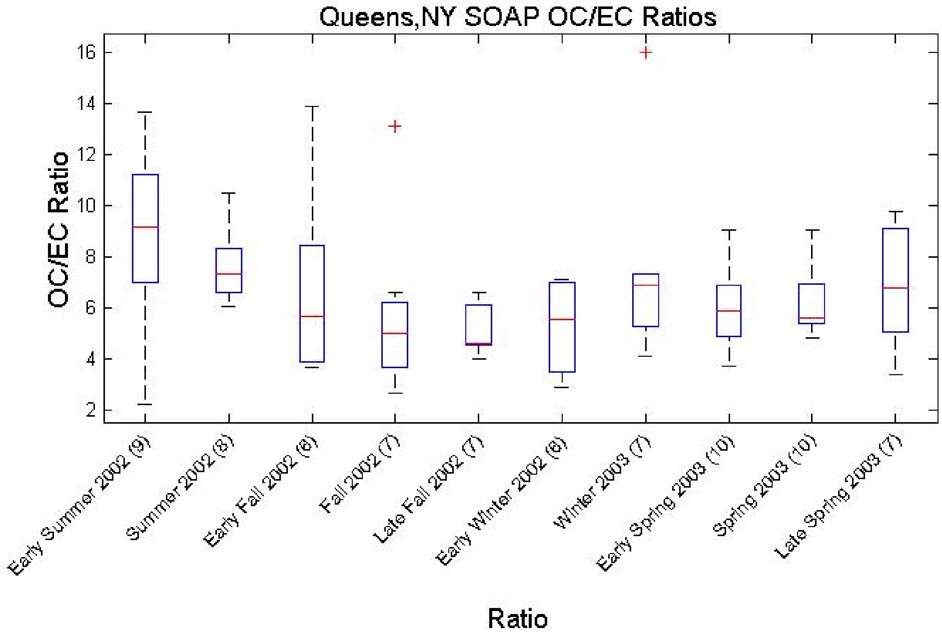


Figure 26- Westport, CT Box-Whisker Plot of OC/EC Ratios based on Ambient Concentrations from (a) SOAP and (b) STN Sampling Studies

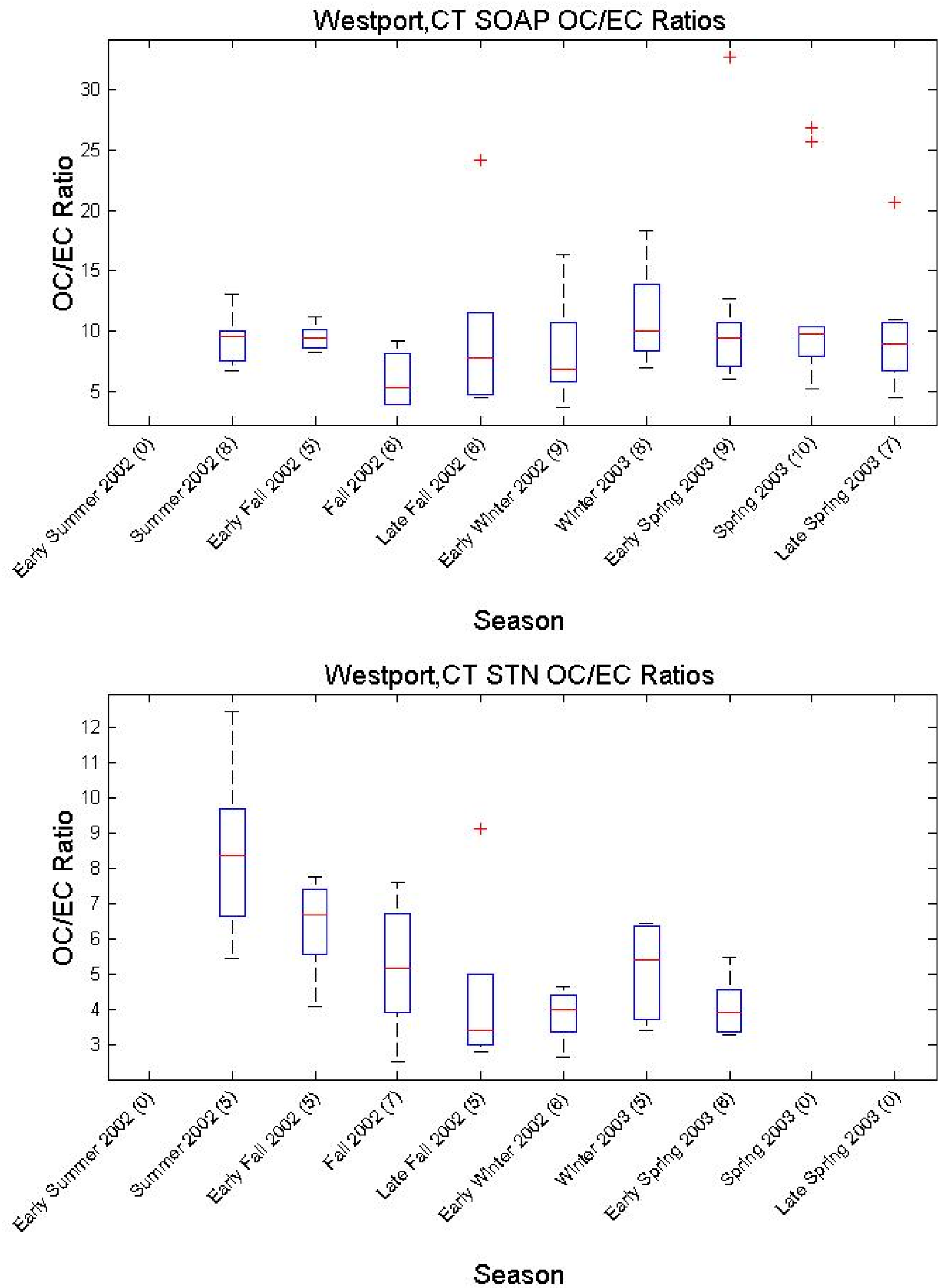


Table 11 compares the OC/EC ratios calculated for the SOAP and STN fine particle filters. Because the STN study did not have successful filters for all the days the SOAP sampler was operating at Chester, the extreme summer outlier I (July 28th, 2002) was not collected. This explains the large discrepancy between the mean, range, and standard deviation for the SOAP and STN OC/EC values. However the extreme outlier for the Elizabeth STN (December 1st, 2002) OC/EC ratios indicates sample heterogeneity collected on that day by the two samplers. However, the mean and median OC/EC ratios are very close. Table 12 shows the same statistical data compared with those extreme values removed. The SOAP OC/EC ratios are consistently higher. Because the SOAP filters per site exceed the STN filters by at least 10 sampling days, it is possible the difference in daily filter sets is because of the missing samples. It is also shown that over all four sites, the highest OC/EC ratios are seen at Chester, followed by Westport, Queens, and the lowest at Elizabeth. This pattern follows the relationship of each site to transportation sources and population density.

Table 11- Comparison of SOAP and STN OC/EC Ratio Statistics

Chester, NJ	**N**	**Mean**	**Median**	**Range**	**StanDev**
SOAP	77	19.88149064	14.28835	217.3004279	25.76322407
STN	58	9.135610224	7.352009	25.18445387	5.846465711
Elizabeth, NJ	**N**	**Mean**	**Median**	**Range**	**StanDev**
SOAP	76	5.268793411	3.996253	24.9413255	4.191915
STN	63	6.138224377	3.462658	142.3618199	17.69480824
Queens, NY	**N**	**Mean**	**Median**	**Range**	**StanDev**
SOAP	77	6.614317082	6.453561	13.80981227	2.704647054
STN	67	4.79037482	4.351157	8.980599432	1.732374552
Westport, CT	**N**	**Mean**	**Median**	**Range**	**StanDev**
SOAP	68	10.04647881	9.271166	28.92105982	5.502927102
STN	39	5.257777928	4.576195	9.926653489	2.16264364

Table 12- Comparion of SOAP and STN OC/EC Ratio Statistics Adjusted for Extreme Values

Adjusted Chester, NJ	N	Mean	Median	Range	StanDev
SOAP	76	17.24547758	14.13405	67.03737831	11.42016964
STN	58	9.135610224	7.352009	25.18445387	5.846465711

Adjusted Elizabeth, NJ	N	Mean	Median	Range	StanDev
SOAP	75	5.235746377	3.914575	24.9413255	4.210165218
STN	62	3.920327538	3.443073	8.707642782	1.804881698

Queens, NY	N	Mean	Median	Range	StanDev
SOAP	77	6.614317082	6.453561	13.80981227	2.704647054
STN	67	4.79037482	4.351157	8.980599432	1.732374552

Westport, CT	N	Mean	Median	Range	StanDev
SOAP	68	10.04647881	9.271166	28.92105982	5.502927102
STN	39	5.257777928	4.576195	9.926653489	2.16264364

Figures 27 and 28 show the seasonal averages of the SOAP and STN OC/EC ratios, respectively. Raw data for the calculated OC/EC ratios are shown in Tables 13 (SOAP) and 14 (STN). A dash indicates that enough data was not collected to generate an average OC/EC ratio for that particular season. The average OC/EC ratios at Chester for both SOAP and STN $PM_{2.5}$ are higher across all seasons, with a summer peak. Queens and Westport also have peak OC/EC ratios in the early summer and summer composites. The average OC/EC ratio at Elizabeth peaks in the late fall for SOAP and winter for STN. Each of the other three sites exhibits OC/EC peaks to varying degrees in these cooler periods. This pattern could indicate an increase of wood and other fuel burning for cold weather heating, causing an increase in organic carbon emissions (Rogge et al. 1998). The higher OC/EC at Chester during warmer months points toward greater secondary OC formed locally, as well as the receiving of aged aerosols from across the Ohio Valley at Chester. Secondary OC mass also occurs at Westport, followed in magnitude by Queens. Secondary organics may be present at Elizabeth as well, but the site exhibits consistently the lowest OC/EC ratios compared to all other sites. The low OC/EC ratios suggest local transportation sources as dominant contributors to $PM_{2.5}$ mass at Elizabeth. A calculated average OC/EC ratio for the warm weather seasons at the Elizabeth site is given in Tables 11 and 12. The average excludes the cooler periods of late fall, early winter, and winter. The impact of possible wood burning would be expected to be at a minimum during the warm seasons. The shaded areas in Tables 11 and 12 indicate the OC/EC ratios higher than the Elizabeth warm weather baseline ratio. Ratios higher that the Elizabeth low OC/EC show the presence of secondary OC in the summer, and the presence of possible wood smoke in the summer. This observation will be useful to examine with additional annual OC and EC measurements.

Figure 27- Seasonal OC/EC Ratios based on SOAP Ambient Concentrations

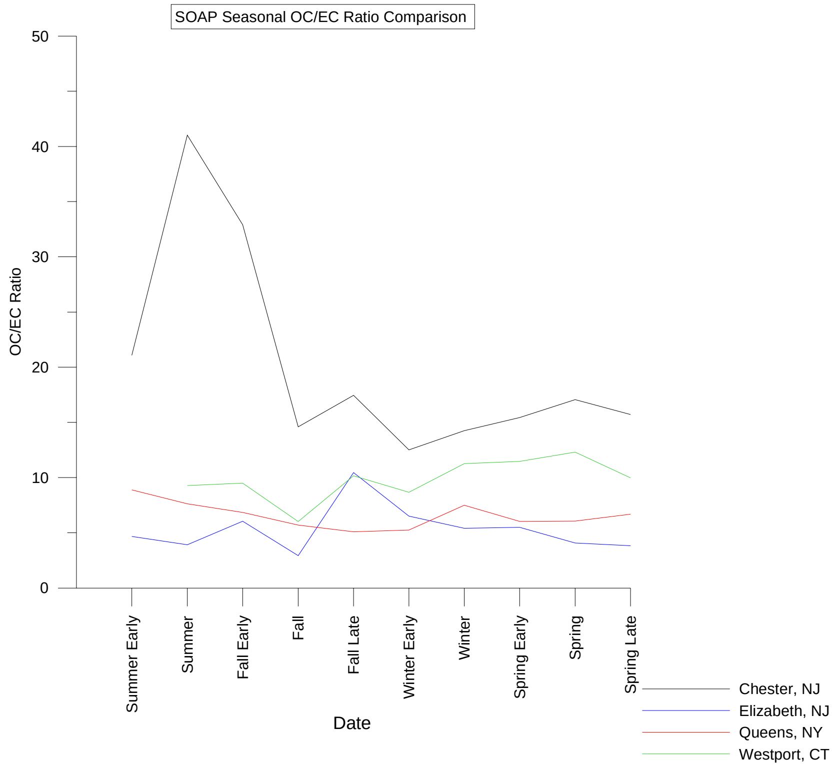


Figure 28- Seasonal OC/EC Ratios based on STN Ambient Concentrations

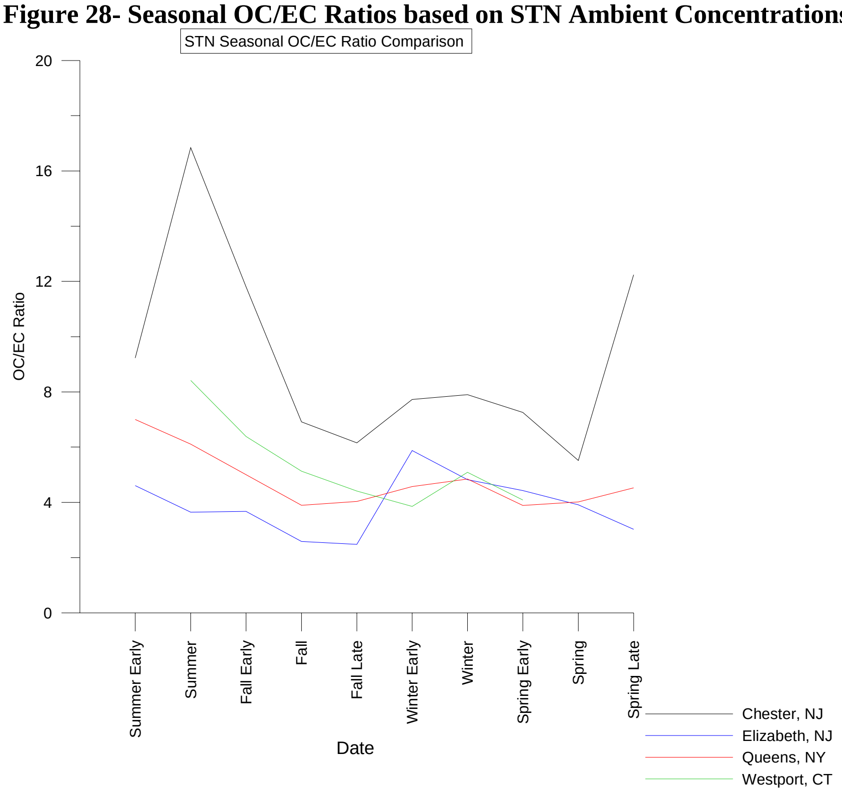


Table 13- SOAP Seasonally Averaged OC/EC Ratios

	Chester, NJ	Elizabeth, NJ	Queens, NY	Westport, CT
Summer Early	21.08	4.67	8.88	-
Summer	41.01	3.91	7.63	9.28
Fall Early	32.91	6.04	6.85	9.50
Fall	14.59	2.92	5.69	6.01
Fall Late	17.46	10.46	5.09	10.16
Winter Early	12.51	6.52	5.26	8.67
Winter	14.25	5.40	7.49	11.28
Spring Early	15.43	5.50	6.02	11.47
Spring	17.07	4.08	6.07	12.31
Spring Late	15.72	3.83	6.68	9.98
Avg. of warm weather OC/EC Ratios at Elizabeth		4.42		

Table 14- STN Seasonally Averaged OC/EC Ratios

	Chester, NJ	Elizabeth, NJ	Queens, NY	Westport, CT
Summer Early	9.23	4.61	7.00	-
Summer	16.84	3.64	6.11	8.42
Fall Early	11.81	3.67	-	6.39
Fall	6.92	2.58	3.90	5.13
Fall Late	6.16	2.48	4.03	4.41
Winter Early	7.73	5.87	4.58	3.85
Winter	7.90	4.82	4.85	5.09
Spring Early	7.26	4.43	3.89	4.09
Spring	5.52	3.91	4.02	-
Spring Late	12.24	3.03	4.53	-
Avg. of warm weather OC/EC Ratios at Elizabeth		3.70		

Conclusions

This study examined the ambient elemental carbon (EC) and organic carbon (OC), and $PM_{2.5}$ mass data at four receptor sites in the NY City metropolitan area. The key objectives of the study were to determine: 1) the trends in these species over the course of an annual cycle; 2) possible differences in the reported EC and OC mass concentrations; and 3) seasonal and temporal patterns of the OC and EC and whether the ratio of OC/EC could be used to understand differences in secondary and primary OC. This study contrasted a site dominated by motor vehicle traffic, Elizabeth, NJ, with the less trafficked sites of Chester, NJ, a regional background site upwind of NY City. Queens, NY was in a densely populated urban residential area and Westport, CT was a suburban site downwind of NY City. All four sites demonstrated peaks in both OC and EC concentrations in the cool winter months by both mass and percentage composition (relative to calculated inorganic composition). Although Chester, NJ did not have the highest OC and EC ambient mass concentrations, the site was unique in that it showed a larger percentage of OC over the entire course of the year, with a large peak in OC mass also in the summer. The seasonal variation in OC and EC ambient mass concentrations were shown by plotting the means, medians and inter-quartile ranges (IQRs) of the ambient data.

As the data provided came from two parallel data sets from collocated samplers, a determination of the difference, if any, between the two protocols was made. Blank levels of OC and EC mass were measured directly by the SOAP network. Whereas the calculation of approximate blank values was necessary from linear least squares regressional analysis of the STN OC and EC ambient mass data relative to the total $PM_{2.5}$

mass data. The SOAP blank measurements showed relatively little in the way of systematic blank concentrations. However, when linear least squares regressional analysis was performed on the SOAP ambient OC ambient mass concentrations, on the order of 1.4 to 1.5 $\mu g/m^3$ OC was overestimated as the y-intercept ambient mass. In general, the OC/EC ratios calculated for the SOAP and STN ambient concentrations indicated the SOAP network generally measured more OC relative to EC. In comparing the two studies using the systematic nature of SOAP measurements consistently lower concentration measurements was shown. One factor that could produce the consistently lower SOAP OC concentrations was the higher face velocity associated with the Tisch sampler.

Although the ambient sample means were shown to be statistically different by one-way ANOVA analysis, the plots of the SOAP and STN ambient OC and EC concentrations were generally consistent in the offset differences. Thus, they both showed the same trends and chemical mass balances. This investigation examined the extent of secondary OC mass to the $PM_{2.5}$ mass. Though winter and summer peaks were noticed for ambient concentrations, it was necessary to determine the chemical mass balance for the total $PM_{2.5}$ and the OC/EC ratios. During the summer months at the Chester, NJ site the OC/EC ratios was 5-7 times greater than any other site. An OC/EC peak was seen in the warm months at Chester, NJ and Westport, CT the downwind site. Elizabeth, NJ showed a fairly constant OC/EC ratio over the course of the year. The Chestet OC/EC ratio data and the chemical mass balance plots show no correlation between the OC and EC components. This observation supports increased OC fine particle mass contributed by local and upwind secondary sources. At the Elizabeth, NJ site, motor vehicle traffic dominates year round and the major sources of OC are primary, though there is a peak in OC/EC ratio in the winter, perhaps due to seasonal wood smoke emissions. Intermediate levels of secondary OC were indicated for Queens, NY and Westport CT in the warmer months.

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Appendices

Appendix 1- Chester, NJ Ambient OC, EC Data from SOAP and STN Networks

	SOAP		STN		
Sample Date	**OC (ug/m3)**	**EC (ug/m3)**	**OC (ug/m3)**	**EC (ug/m3)**	**PM2.5 (ug/m3)**
05/26/02	3.142706596	0.132158483	4.920767677	0.450474626	17.92003315
06/01/02	4.02218665	0.116034574	4.237598084	0.368885918	9.382410558
06/04/02	2.799270044	0.246230365	3.431438532	0.46214521	4.745692768
06/07/02	1.660691674	0.118994889	1.876389751	0.206466723	3.093102382
06/10/02	4.185071593	0.225321162			
07/10/02	4.074131234	0.118639088			
07/13/02	3.648769815	0.175479771	2.916203608	0.790018762	6.291903043
07/16/02	4.67007012	0.259964014			
07/19/02	4.56052048	0.319177563	4.07140803	0.318958748	8.665153703
07/25/02	2.289272728	0.219952881	6.597687205	0.624723945	11.04801239
07/28/02	6.605846761	0.029996786			
08/12/02	4.481664259	0.27303545	9.932332454	0.377580199	
08/15/02	3.498512882	0.169998664	4.308068271	0.271458584	
08/18/02	3.395588503	0.153801281	4.591946392	0.170093103	
08/21/02	3.330090506	0.204906937	4.282076484	0.298393409	
08/24/02	2.13343549	0.291309188	3.21195076	0.420143047	
08/27/02	3.564974623	0.240245497	6.887047757	0.425592506	
09/02/02	1.499647081	0.038624909	2.68006558	0.189814981	
09/05/02	1.656034333	0.023672705	2.027209652	0.26842998	
09/08/02	3.99757545	0.197209219	6.747114338	0.266614356	
09/11/02	2.152073917	0.079637743	2.687732518	0.304108136	5.054672994
09/20/02	2.526716257	0.297377917	3.353575593	0.429024823	9.501187648
09/26/02	0.737322404	BDL	2.492684893	0.345308893	7.544439851
10/05/02	1.941123764	0.283728687	2.762361084	0.478118735	15.40529363
10/17/02	1.77751732	0.174295847	1.603462664	0.386075102	4.648280136
10/20/02	5.098603354	0.165905453	2.150650875	0.436014898	6.817477533
10/23/02	1.890764042	0.156241592	2.398834046	0.191413111	7.757550683

10/26/02	1.613367136	0.082017511	1.475149843	0.216721107	4.952538176
10/29/02	1.407869718	0.141877902	1.488612541	0.529819329	6.000413822
11/01/02	1.939139114	0.251933882	3.183995723	0.520773902	8.46495303
11/04/02	2.120329617	0.241537437			
11/13/02	1.322294475	0.104538728	1.646643812	0.144320612	5.373010953
11/16/02	1.755616455	0.153593021	2.459388103	0.457186167	6.101344364
11/19/02	2.909787262	0.174401737			
11/22/02	1.905501832	0.356876686	2.38321631	0.869575715	8.67589341
11/28/02	1.756376645	0.104256855			
12/01/02	0.921161199	0.016616324	1.18252333	0.113598506	2.168077638
12/04/02	1.930844292	0.148064721	2.546654779	0.383659875	5.364696172
12/07/02	3.894390355	0.24352864	7.359745421	0.52141276	21.2061653
12/13/02	2.548850303	0.447968797	2.399449486	0.730889149	11.34020619
12/19/02	2.811306563	0.459930726	4.100306691	1.040201521	14.63163318
01/09/03	1.567415541	0.055738529	2.589319584	0.395722366	7.41885626
01/12/03	1.911515735	0.188549813	1.892871414	0.261307685	5.560704356
01/24/03	1.223085304	0.045771777			
01/30/03	3.458210605	0.344659653	7.978807406	0.469900907	
02/02/03	1.518361143	0.062984455	3.50550461	0.314537005	
02/08/03	3.564915369	0.886806948	3.152183705	0.385714527	
02/11/03	7.602068807	2.605182799	1.830139841	0.50037023	
02/14/03	2.394323269	0.171270876	2.832040536	0.376381404	
02/17/03	2.016891822	0.100039316			
02/26/03	3.065519586	0.381539471	2.993813886	0.661244933	
03/01/03	3.492479461	0.30617148			
03/04/03	2.648363063	0.27623529			
03/10/03	1.592131306	0.129837062			
03/16/03	1.707385683	0.113474919	3.569447688	0.234370439	9.389186958
03/19/03	1.736098982	0.229923711	1.842528213	0.43403925	6.086866811
03/22/03	1.822706285	0.168807839	2.023922079	0.369805239	9.690721649
03/25/03	4.666521869	0.376175966	5.086866279	0.772605179	19.6765221
03/28/03	1.75787646	0.141257063			

03/31/03	1.012215318	0.068562285			
04/03/03	1.121184197	0.124788032			
04/06/03	1.303095927	0.032901277	4.476707649	0.438716297	11.13057817
04/09/03	1.333419184	0.127394311	0.974562973	0.537775827	6.627316972
04/12/03	1.669688478	0.066217163			
04/15/03	1.528830576	0.270494652	4.606295284	0.63282015	16.8371036
04/18/03	5.959315669	0.523006989			
04/24/03	1.101872444	0.030537521	1.606740414	0.27434234	4.952538176
04/27/03	1.617233963	0.081021561	2.029881577	0.361171115	7.016095749
04/30/03	2.042895484	0.137513902	2.093751564	0.437915682	3.917525773
05/03/03	1.884448295	0.054173708	1.402026958	0.220715459	4.427512356
05/06/03	2.018831161	0.269602326	3.01327852	0.649170107	13.39101772
05/09/03	2.407383831	0.262431721	3.006438715	0.734717026	16.92990606
05/12/03	2.93598681	0.140483314			
05/15/03	2.298565103	0.221294211			
05/18/03	2.733629141	0.129973555			
05/21/03	2.416631591	0.113224101	2.667672156	0.297258826	8.563763929
05/24/03	1.03614998	0.076604999	1.672210546	0.370582218	5.470120755
05/30/03	5.257427676	0.292953199	5.30036491	0.228182669	16.21900826

Appendix 2- Elizabeth, NJ Ambient OC, EC Data from SOAP and STN Networks

	SOAP		STN		
Sample Date	**OC (ug/m3)**	**EC (ug/m3)**	**OC (ug/m3)**	**EC (ug/m3)**	**PM2.5 (ug/m3)**
05/26/02	4.487579172	0.691715177	2.990160643	0.689879426	13.19723683
06/01/02	7.590688611	0.965408378	6.738999699	1.024707388	18.46122112
06/04/02	7.045094459	2.590097852	5.324514299	2.560785548	11.76592012
06/07/02	3.43759171	1.403026948	3.724330207	1.674444571	6.811145511
06/10/02	6.693479501	2.13666168			
07/10/02	6.72904915	1.008388575	8.574340863	1.307718802	20.81830362
07/13/02	6.112932636	1.372861237	6.399881602	1.13001568	12.68564356
07/16/02	4.780079847	1.06476185	9.778654448	1.433914454	15.5943406
07/19/02	9.798464145	2.604384965	8.878174016	3.38293569	44.30902706
07/25/02	3.264324124	1.928322525	5.443101902	2.286686788	8.370362716
07/28/02	5.569949132	0.99519626	6.698475856	1.356544092	27.96697626
08/12/02	8.14688569	3.276845826	10.39098686	3.830338755	36.11971104
08/15/02	9.8764943	2.044298403	8.272598873	4.018	20.42921998
08/18/02	5.258344366	0.670098729	6.588059306	1.022240306	26.34025411
08/21/02	5.077495094	1.891515867	5.873824968	2.259528714	13.10223873
08/24/02	3.116201021	0.972459569	6.508860955	1.18383765	10.74047299
08/27/02	6.928803839	2.358727479	8.026030023	3.18039402	18.79388682
09/02/02	1.895750764	0.297670561	2.691441884	0.535369143	4.751084487
09/05/02	2.549661034	1.38697512	6.891936828	1.454198563	8.573494474
09/08/02	5.141029006	0.769977468	6.295368297	1.270046019	14.03074384
09/11/02	2.958291646	1.390936753	3.132407881	1.620307955	7.739139408
09/20/02	4.425003777	3.776461545	4.972280087	3.866444078	15.89103292
09/26/02	4.345551533	2.372729477	4.857656558	2.614577309	13.61667011
10/05/02	2.3816855	0.13209041	3.311400173	0.813079214	17.93259817
10/17/02	3.202262842	1.993971445	5.41991185	2.14841289	12.38645747
10/20/02	3.28617689	0.527942823	3.607855642	0.944183002	7.533539732

10/23/02	3.97163627	1.632243274	5.054216198	1.88285974	12.18756455
10/26/02	2.735602987	0.884336417	3.236139065	1.46183829	10.31140441
10/29/02	3.367150908	1.190560249	4.002314331	1.408492041	9.48942754
11/01/02	4.195015758	1.676610356	5.113837761	2.03249065	14.24589656
11/04/02	4.344749865	1.682498082			
11/13/02	2.31068144	0.944259338	3.373332995	1.591167941	8.152734778
11/16/02	2.564085825	0.112651641	4.447556403	1.611907565	11.46575767
11/19/02	2.989772441	0.114493385	9.199620563	3.575369935	20.22495099
11/22/02	6.334335838	2.445292607	7.757691737	3.718849284	27.33085809
11/28/02	2.831235674	0.316811668			
12/01/02	1.834051975	0.236733704	3.590838898	0.024997516	3.719392499
12/04/02	4.419737168	1.229757271	5.994853174	1.623874332	14.2194745
12/07/02	6.958866379	0.86385276	10.54310832	1.319411051	37.80991736
12/13/02	9.879233913	3.118049737	11.14472131	4.651415965	31.5245478
12/19/02	8.453997631	2.989232536			
01/09/03	5.153353776	1.719215859	6.95067563	2.030290659	16.50675745
01/12/03	3.039900377	0.323886316	3.216644741	0.421157717	9.583676834
01/24/03	2.18122639	0.557206484			
01/30/03	11.22522708	1.964281505	12.62152961	3.368985562	44.06150036
02/02/03	2.115351957	0.175226757	3.388639637	0.443133136	9.187570971
02/08/03	3.713352866	0.419223432	6.531766466	0.653591591	19.11354479
02/11/03	3.259917391	0.780198383	4.250050391	1.27337421	10.84262701
02/14/03	4.733060292	0.922795555	6.107121897	1.226974149	15.48786784
02/17/03	3.720024135	0.564894477			
02/26/03	6.284827627	1.074830448	7.24116573	2.091215906	27.7892562
03/01/03	4.975345196	0.823099109	5.158648742	1.198853778	31.24677735
03/04/03	6.736612711	1.872324639	7.830555868	2.747894538	19.59570957
03/10/03	2.783475796	0.514151698	4.617095614	0.773432539	9.274525969
03/13/03	6.200000723	1.227222955			
03/16/03	4.461071854	0.781047105	5.69630295	1.310060844	12.67649181
03/19/03	3.143781528	1.044616985	12.45275192	1.520939356	13.41589267
03/22/03	2.870249706	0.431369395	3.511373919	0.759899391	13.73257615

03/25/03	6.511267395	1.59670848	7.368175207	2.592319059	25.80245639
04/06/03	1.743758743	0.204556783	2.408882899	0.506069612	5.675954592
04/09/03	4.260292425	1.122104795	4.59020771	1.839751669	17.63250155
04/12/03	2.188985488	0.268579188	2.397446576	0.605312814	4.535614885
04/15/03	5.694127465	1.496127411	6.208480223	2.11507787	
04/18/03	4.290591981	0.986531278			
04/24/03	2.114797761	0.543755893	2.321157621	0.7840155	6.49618478
04/27/03	2.902737814	0.431129941	3.917388418	0.751301509	6.702412869
04/30/03	4.336158078	1.543942013			
05/03/03	2.746476041	0.358855411	3.225708175	0.596977694	8.052859798
05/06/03	5.742140392	1.573001423	6.232385369	2.454862416	24.75502837
05/09/03	4.398304756	1.710788589	10.31810471	2.330483843	20.62068254
05/12/03	3.509032157	1.948905867			
05/15/03	5.126257406	1.466507183			
05/21/03	4.820504815	1.579431146	5.193775271	2.260319132	14.12808085
05/24/03	3.317211191	0.720087244	3.897364613	1.249253937	15.14371072
05/27/03	4.381703262	2.333064003			
05/30/03	5.763970153	1.26604633	7.073922663	1.930659369	21.26341866

Appendix 3- Queens, NY Ambient OC, EC Data from SOAP and STN Networks

Sample Date	SOAP OC (ug/m3)	SOAP EC (ug/m3)	STN OC (ug/m3)	STN EC (ug/m3)	STN PM2.5 (ug/m3)
05/26/02	2.564018554	0.192869408	3.072590365	0.448422358	12.24879607
06/01/02	5.599753744	0.612408976			
06/04/02	2.817635555	0.448504521	1.107989687	0.163353238	7.225313292
06/07/02	1.975540755	0.238801589	2.240715217	0.516875418	4.692106429
06/10/02	3.847747234	1.753962928	4.757886345	0.941610125	16.23484496
07/10/02	7.8952387	0.578310213	5.169450585	0.496700235	23.74922241
07/13/02	3.127781983	0.336002687	1.84517708	0.209733567	10.00705885
07/16/02	5.116159878	0.485166529	5.209124822	0.744351334	11.29521229
07/19/02	6.944687317	0.960140724	7.917641657	1.169161816	40.74089376
07/25/02	3.420755642	0.325396309	3.185712625	0.44828588	4.48523601
07/28/02	6.024438973	0.73726071	0.990118101	0.177494912	22.58667306
08/12/02	5.454934209	0.843375029	3.426732926	0.531749363	23.09161975
08/15/02	2.402698257	0.359871438	3.574763703	0.529165178	9.38538209
08/18/02	5.147745587	0.604231268	6.818277848	0.980486408	25.74643977
08/21/02	3.59310619	0.481318524	3.928399899	0.679272865	8.802524631
08/24/02	2.719412087	0.377080456	3.262314059	0.560119225	7.639927039
08/27/02	5.821132548	0.961904393	5.97832773	1.35405627	16.6092263
09/02/02	1.465229045	0.105555731			
09/05/02	3.091537887	0.801615111			
09/08/02	4.391054891	0.520862354			
09/11/02	4.010917989	1.043792662			
09/20/02	2.410366732	0.656248351			
09/26/02	2.601040268	0.504649495	2.553738306	0.574659899	5.976591717
10/05/02	3.187345548	0.428503607			
10/17/02	3.03987319	1.156120255	0.745247375	0.386465196	8.050460762
10/20/02	2.882683224	0.437846174	2.535962845	0.357637519	7.512243755

10/23/02	3.299349336	0.663244819			
10/26/02	1.474278131	0.112615357	2.011979241	0.622656965	6.143882921
10/29/02	2.233415657	0.588574697			
11/01/02	4.590771238	1.149559127	3.837782086	0.996378864	14.19087114
11/04/02	5.09690591	1.122725216	4.097251361	1.412227774	19.46300359
11/13/02	2.63448724	0.729282373	2.946731314	1.054076298	7.636121965
11/16/02	3.020628981	0.468056151	3.436072242	0.580750387	9.253111887
11/19/02	5.255106325	1.070897745	1.278205513	0.368059729	17.17771059
11/22/02	3.906018023	0.863457155	3.773748877	1.213529288	15.60230655
11/28/02	1.895445782	0.287309598	2.458205817	0.491183441	10.20619813
12/01/02	1.216165866	0.264701696	1.845379785	0.463554774	3.692486314
12/04/02	5.782795403	0.892352397	4.870478606	0.950128948	14.76933283
12/07/02	3.507125582	0.755050066			
12/13/02	5.388545664	1.541776813	4.458452911	1.446140189	18.38098017
12/19/02	5.825102546	2.038175752	5.089353865	2.170010046	20.24476288
01/09/03	3.409807998	0.835376476	3.490659511	1.057416604	13.15407228
01/12/03	2.079042683	0.293405479	3.786557767	0.466196367	11.20053057
01/24/03	3.533277378	0.689808602	2.925247002	0.714360018	7.801801015
01/30/03	2.524508882	0.366390391	0.717813832	0.09818	28.81758731
02/02/03	5.417681058	0.774388732	3.206703626	0.762761172	10.49923222
02/08/03	2.979769719	0.407313089	3.507483647	0.516819764	17.01315342
02/11/03	3.038072226	0.531265213	3.225918092	0.543058531	12.86200354
02/14/03			1.194701474	0.19580946	12.07368719
02/17/03	3.107377099	0.194167901	0.403784699	0.282963142	7.470429763
02/26/03	2.242399913	0.307201937	0.885332042	0.232858177	10.25364244
03/01/03	3.799737793	0.645738487	4.001803957	0.849339958	22.32643056
03/04/03	2.904390993	0.783177101	3.088776736	1.069959611	11.78227676
03/10/03	2.418025796	0.35142186	2.77635036	0.638071728	9.166666667
03/16/03	3.87733226	0.800127843	3.830062956	1.046980598	11.66362324
03/19/03	2.340667299	0.259232161	2.602440624	0.643387902	11.11848652
03/22/03	3.915181434	0.669745298	5.647422144	1.04244674	15.10749573
03/25/03	3.839029969	0.669138471	3.86175438	1.07329283	17.54677039

03/28/03	3.158413359	0.93914644		3.216893612	1.112260908	12.73541809
03/31/03	1.690791858	0.337273644		2.160156739	0.533387881	5.020746808
04/03/03	2.999996493	0.307295174		3.047405553	0.770859147	18.00531025
04/06/03	1.925574198	0.256595127		2.071272624	0.468716919	6.762363068
04/09/03	1.778030736	0.274569529		1.59456904	0.540046855	8.960424456
04/12/03	2.243178576	0.520072537		2.504395281	0.87738298	4.937554493
04/15/03	5.449545608	0.979448718		4.42976403	1.167146187	15.51674024
04/18/03	1.854580943	0.354264295		2.139865427	0.424401669	8.670400497
04/24/03	1.620647325	0.302206815		2.768698228	0.686032106	9.169363811
04/27/03	3.08617229	0.554789978		3.615676116	0.875116236	9.04451704
04/30/03	2.384926678	0.345188928				
05/03/03	2.627631587	0.291026513		2.57044536	0.490054572	7.625
05/06/03	2.994339159	0.526507489		3.148353079	0.899197664	12.32365126
05/09/03	3.615664423	0.643348471		5.347115582	1.848029755	21.52990876
05/12/03	5.265451325	1.087562326		3.144894974	0.756148537	10.20746872
05/15/03	3.398448017	0.490366564		2.860839976	0.858507705	8.255205864
05/18/03	3.145416233	0.443775029		3.077246644	0.487028535	8.088601361
05/21/03	6.143430865	1.211849408		5.141968809	1.254318223	15.43312307
05/24/03	1.693136911	0.173873298		2.109116157	0.355197951	8.380351777
05/30/03	7.481173826	1.110530804		5.471707076	1.232115595	21.07668844

Appendix 4- Westport, CT Ambient OC, EC Data from SOAP and STN Networks

	SOAP		STN		
Composite	**OC (ug/m3)**	**EC (ug/m3)**	**OC (ug/m3)**	**EC (ug/m3)**	**PM2.5 (ug/m3)**
07/25/02	2.089109538	0.214671599			
07/28/02	2.575958872	0.253991275	3.587911793	0.658965877	19.81628651
08/12/02	5.258476815	0.741019138			
08/15/02	2.567657429	0.377165851	3.21645892	0.258328324	11.45983894
08/18/02	4.816367512	0.367891949			
08/21/02	2.808823136	0.3002268	3.115834966	0.372563313	7.83990097
08/24/02	1.924905677	0.192973904	2.690967636	0.307297874	6.096300889
08/27/02	4.252369857	0.531284732	5.426688427	0.767277545	13.64623178
09/05/02	2.37789884	0.243670884	1.991354334	0.256055727	4.438480595
09/08/02	4.350431249	0.492811302	5.134839331	0.702716954	10.73492981
09/14/02	3.071176224	0.370917025	2.818833751	0.687276499	10.95041322
09/26/02	5.346506904	0	3.699260993	0.516412927	9.090909091
09/29/02	3.476106617	0.309985292	3.163554128	0.47281647	8.042895442
10/17/02	2.05991489	0.523511343	2.11044977	0.836021446	5.25990099
10/20/02	2.308272602	0.251263514	2.687730402	0.520875765	8.865065457
10/23/02	3.649881694	0.908485089	3.699240446	0.915544866	10.72717896
10/26/02	1.856370405	0.431742345	1.5114348	0.274965494	4.330789854
10/29/02	2.227891469	0.342215159	2.752634185	0.709283927	6.081220367
11/01/02	3.015257459	0.654815457	3.262892632	0.960878891	9.920429885
11/10/02	3.332392338	0.353794796	4.676014162	0.772290902	15.17497677
11/13/02	1.243420129	0.152940011	1.91620644	0.251409503	4.120737612
11/16/02	2.175995682	0.205302055	2.597283647	0.714862854	7.745533409
11/19/02	3.644653426	0.752928086	3.520643781	1.258303011	13.10088715
11/22/02	2.60018074	0.503510778	3.52047496	1.134843158	10.21355617
11/28/02	2.975324646	0.256395512			
12/01/02	2.033968042	0.084152387	3.751031262	0.411510733	8.053691275

12/04/02	6.75056173	0.73657511	5.00477411	1.168934652	13.3085732
12/07/02	4.156065162	0.675821591	7.043912322	2.084159628	23.60338075
12/10/02	6.249972508	1.167331404			
12/13/02	3.8719576	1.034240268	4.708091965	1.775262312	17.13813752
12/19/02	6.230126003	0.904203486	5.230505101	1.40940785	17.22359736
01/03/03	1.903959493	0.20330761	2.285553342	0.489769682	6.181106418
01/06/03	5.05727564	0.335988745			
01/09/03	3.408993433	0.48331398	3.271480149	0.954362448	10.72054427
01/24/03	1.550597249	0.089717007	1.932471515	0.504511758	5.680057833
01/30/03	4.689223008	0.524253408	7.320362179	1.138111736	22.7014756
02/05/03	1.350052047	0.082633854	1.369428285	0.309832219	4.857881137
02/08/03	2.823118963	0.27239795	3.355114371	0.62266462	14.89141675
02/11/03	2.266225478	0.287390287	2.951852273	0.463669264	9.303287161
02/14/03	3.803373182	0.358978101			
02/17/03	2.255047093	0.123084824			
02/20/03	4.832052316	0.805664927			
02/26/03	3.537229948	0.361725033			
03/01/03	5.582897809	0.439988718			
03/04/03	2.584664636	0.312841739			
03/10/03	3.879768632	0.548148684			
03/16/03	5.529213954	0.582102798	6.368413333	1.158569697	16.20059849
03/19/03	1.712748492	0.174867401	1.868890397	0.571324437	8.165374677
03/22/03	2.383175557	0.336399303	2.67215222	0.681994283	8.255933953
03/25/03	2.970591321	0.489160926	3.890212128	0.850097586	16.49994844
03/28/03	4.238211707	0.469906609			
03/31/03	1.151955765	0.055701025			
04/03/03	3.108285772	0.303736641			
04/06/03	1.393315963	0.042654928	1.236390273	0.31474741	5.468991848
04/09/03	1.961665629	0.19533248	1.996769527	0.59515179	8.76108019
04/12/03	6.08556091	0			
04/15/03	4.851027443	0.611415256			
04/18/03	1.205659138	0.115651133			

04/24/03	1.374713089	0.137556913
04/27/03	2.068941669	0.21318325
04/30/03	3.32810237	0.124161986
05/03/03	2.033983379	0.0791237
05/06/03	1.621664397	0.168624257
05/09/03	4.096699923	0.782757794
05/12/03	1.390669763	0.140202417
05/15/03	2.392842275	0.308783976
05/18/03	2.200745954	0.277823951
05/21/03	1.292006422	0.117733203
05/24/03	3.491360611	0.765947867
05/30/03	4.715817464	0.730727467