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**SEASONAL VARIATIONS OF FINE PARTICULATE MATTER DERIVED
FROM BIOGENIC AND ANTHROPOGENIC SOURCES**

by

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**A thesis presented to the faculty of Towson University in partial fulfillment of the
requirements for the degree of Master of Science**

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Thesis Approval Page

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Abstract

Seasonal Variations of Fine Particulate Matter Derived from Biogenic and Anthropogenic Sources

Larry Edward Meade

Organic aerosols are classified as solid or liquid particles suspended in the gas phase. Studies have shown that they impact both humans as well as the environment. Organosulfates (OSs) are an important class of secondary organic aerosols (SOAs). In this study, air filter samples were collected between August 2012 and June 2013 in Towson, MD. This particular data set provides a unique insight into the impacts of seasonal variations of OS. Ultra-performance liquid chromatography/electrospray ionization (UPLC-ESI) was used to analyze these samples in order to identify and quantify OS. Seasonal trends show OS derived from biogenic compounds dominate in warmer seasons. During colder months, anthropogenically derived OSs dominate. Many biogenic OSs correlate positively to temperature, while anthropogenic OSs correlate negatively. Meteorological data and air mass back-trajectory analyses provides insight into aerosol origin, as well as meteorological and transport conditions that promote the formation of OSs within the mid-Atlantic U.S. region.

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I. Introduction:

IA. Aerosols

Aerosols are liquid or solid particles originating from various sources that are suspended in the atmosphere, and approximately 300 Tg/yr of primary aerosols enter the atmosphere every year.¹ Primary organic aerosols (POAs) are emitted directly to the atmosphere in particle form. SOAs are called “secondary” because they are not directly released into the atmosphere. Rather, SOAs form from the oxidation of volatile organic compounds (VOCs), which then condense and form an aerosol.²⁻⁵ This study focuses on SOAs. An overview of the process of SOA formation can be seen in Figure 1.

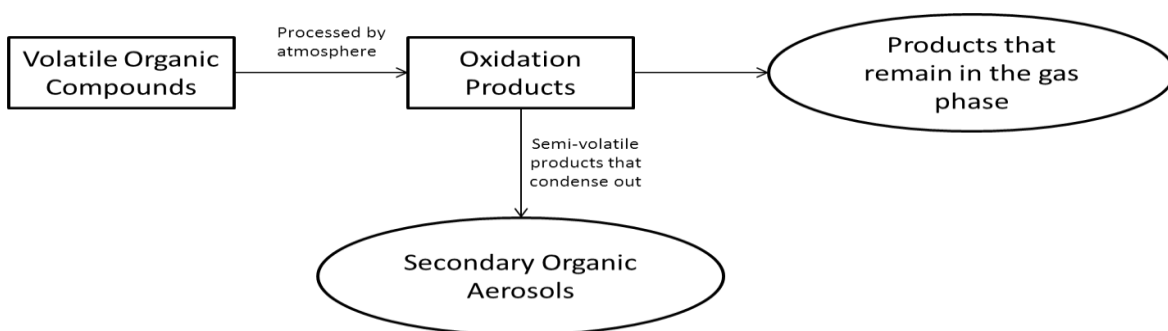


Figure 1- The process of secondary organic aerosol formation.

SOAs can be biogenic or anthropogenic in nature.⁶⁻⁸ Biogenic sources of aerosols include VOCs such as isoprene as well as monoterpenes and sesquiterpenes.^{3,9-12} Isoprene is produced primarily from broadleaf plant species and terpenes are typically produced from coniferous plants.¹³ Monoterpenes consist of two isoprene molecules, and sesquiterpenes consist of three isoprene molecules.¹³ An example of the isoprene structure can be seen in Figure 2.

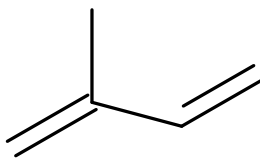


Figure 2- The structure of isoprene.

Yearly global production of isoprene emissions by vegetation is around 600 Tg, as well as approximately 127 Tg/year of monoterpene and sesquiterpenes.¹⁴ Alternatively, anthropogenic aerosols are produced from human activities and can form from vehicle exhaust, chemical synthesis, manufacturing, and other processes.^{8,15,16} The total emission of anthropogenic VOCs into the atmosphere is estimated to be 109 Tg/yr.¹⁷ The percentage of SOA that is biogenic and anthropogenic in nature fluctuates throughout the year, especially in locations with distinct seasons. During summer months when vegetation is actively growing, there is an increase in the biogenic portion of the aerosol mass.¹⁸ This portion then shrinks in the winter months as plants begin to go dormant.^{8,16,19} To understand the potential impacts that these fluctuations can have on the secondary organic aerosol mass of a region as a whole, it is important to have data that encompass a longer period of time, rather than the typical sampling period of a few weeks or a few months.^{10,12,20}

IB. Climate Impacts:

Aerosols are known to alter the energy balance of the Earth in several ways. For example, aerosols can impact the Earth's energy budget through a process called radiative forcing. Radiative forcing is defined as any external disturbance in the radiation

budget, such as increased fossil fuel burning.¹⁷ The formation of SOAs are complex in nature, and as of 2014 SOAs are a new category in the section of the Intergovernmental Panel on Climate Change (IPCC) assessment report detailing the potential contributors of climate warming and climate cooling.^{17,21} Specifically, the IPCC states that radiative forcing occurs when there is some change in the net amount of irradiance occurring in the atmosphere, which forces a change in temperature in order to regain relative equilibrium of the system.¹⁷ That is, the system is forced to change in order to return to a point where there is no net change. Radiative forcing is categorized as either positive or negative; positive forcing denotes a warming effect and negative forcing causing a cooling effect.¹⁷

Aerosols have the potential to cause both positive and negative radiative forcing. They are able to reflect and scatter radiative energy before it reaches the surface of the earth.²² Since less energy is reaching the surface of the earth, a cooling effect can be observed. Some studies refer to these aerosols as “white”.^{7,8} Aerosols are also able to absorb a portion of the radiative energy either directly from the sun or from the energy that is reflected from the surface of the earth, resulting in a positive forcing.^{8,23} Aerosols are also able to re-radiate infra-red (IR) energy.¹⁷ While the majority of radiative absorption from primary organic aerosols occurs in the visible spectrum (i.e. black carbon), recent studies have shown that some classes of SOAs (brown carbon) can absorb radiation in the ultraviolet to near visible spectrum, typically from 300-400 nm.⁷ Warming effects includes factors such as black carbon, biomass burning, and mineral dust.¹⁷ The effective radiative forcing due to SOA specifically is calculated to be -0.12 (-0.4 to $+0.1$) W/m^2 .¹⁷ In comparison, the radiative forcing for anthropogenic CO_2 is estimated to be 1.82 (1.63 to 2.01) W/m^2 .¹⁷ The large relative uncertainties for SOA arise

because we know little about the absorptive and scattering properties of these particles and the connection that these properties have to the aerosol's chemical and physical properties such as particle number, size distribution, chemical composition of the aerosol, and the general morphology.^{4,24} To obtain a better understanding of the potential impacts that these secondary organic aerosols have on the environment it is necessary to analyze data samples from the field and determine the concentration and composition of the observed compounds.

IC. Organosulfates:

Atmospheric OSs are a class of SOA that have the potential to contribute to a variety of environmental and health concerns, so it is important to understand their contributions to the SOA mass.^{25,26} These compounds have an SO_4 group bonded to the structure through an oxygen atom. OSs have been observed in air aerosol samples collected throughout the world.²⁷⁻³² Formation of OS seems to be more common in urbanized areas where the combination of natural sources and human activity can lead to an increased concentration of atmospheric sulfur in the form of SO_3^- or sulfuric acid.⁹ OS forms from sulfuric compounds reacting with either anthropogenic or biogenic volatile organic compounds.^{26,33,34}

While there is a great deal of variety in the types of OSs identified from field measurements, these compounds serve as tracers for different reactions that lead to the formation of SOAs.¹ OS concentrations in the atmosphere can range from as low as 4% to over 30% of the total SOA mass.^{29,35} Even at the lower end of this range, studies have shown that OSs have the potential to impact the degree of radiation absorption that can occur in the UV-visible spectrum.¹² This is especially true in urban areas where OS could

potentially make up 30% of the organic mass in aerosols. Sulfur is a trace element in the atmosphere, and can originate from anthropogenic activities including chemical synthesis and the burning of fossil fuels.^{3,5,17,36} In previous studies, it has been shown that OSs are involved in a variety of reactions that contribute to SOA formation.^{34,37,38} A few OS precursors have been identified from laboratory studies that focused on SOA formation from a variety of sources.^{34,37,38} The sources for the formation of these SOAs include the hydroxyl radical (OH), the nitrate radical (NO₃), or ozone (O₃).^{29,34} These sources initiate the oxidation of different types of biogenic VOCs. Biogenic VOCs include compounds such as isoprene, monoterpenes^{20,30,37,39-41}, and sesquiterpenes.^{28,30-34} Unsaturated aldehydes can also be oxidized this way. This is important because this formation begins in the presence of acidified sulfate aerosol.^{20,33}

Studies have shown that the acidity of the aerosol derived from the presence of sulfate plays a key role in the formation of SOA and OS formation from biogenic precursors.^{33,42,43} This emphasizes the importance of the chemical interactions between biogenic compounds and anthropogenic pollutants, as atmospheric sulfur originates primarily from anthropogenic activity. The SOAs that are formed in these mostly acidic conditions are light-absorbing in nature and in some cases have peak absorbance values at around 400 nm.⁴⁴ This holds especially true from SOAs derived from reactive uptake onto seed aerosol acidified with H₂SO₄.⁴³ Seed aerosol is aerosol refers to the aerosol used in chamber experiments, and are the particles that the rest of the SOA in the chamber reacts with and bonds to.

One particular class of compound observed reacting with seed aerosol are epoxides. Epoxides are cyclic ethers with three-atom rings. This ring approximates to an equilateral

triangle.^{4,45,46} These reactions were observed in laboratory settings where the epoxide and seed aerosol are allowed to react in low NO_x environments. One potential mechanism for the formation of these compounds is ring-opening reactions of epoxides by H_2SO_4 , resulting in the addition of HSO_4^- to the compound.⁴³ This mechanism can be seen in Figure 3.

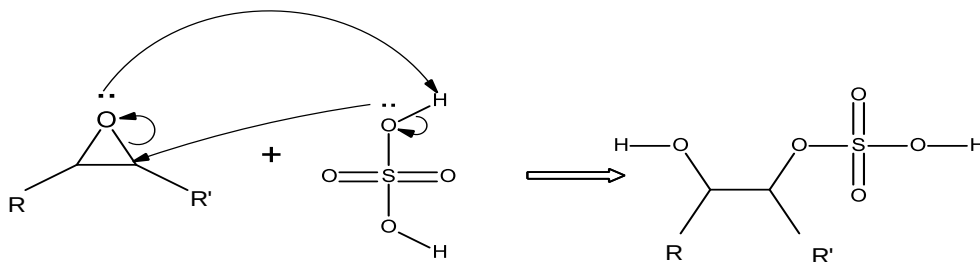


Figure 3-The opening of an epoxide by H_2SO_4

These reactions have been shown to cause a “browning effect”, in which the mixture of H_2SO_4 and isoprene epoxide has clearly led to an increase in light-absorption, which does not occur in mixtures of the SOA and other acids such as HNO_3 and H_3PO_4 .⁴⁷ This indicates that these specific compounds may absorb radiative energy at the edge of the UV-Visible spectrum and thus cause a disturbance through radiative forcing.⁴⁸ These OSs form when acidified aerosols oxidize biogenic volatile organic compounds as well as polycyclic aromatic hydrocarbons that are released from anthropogenic processes.^{29,34} This oxidation allows sulfate particles to bond to the volatile organic compound, forming OSs.^{9,12,13}

OSs have been observed forming from nucleophilic substitution reactions of an organic nitrate group by sulfate in environments where there is an abundance of nitrogen

containing species.^{49,50} In these reactions, one of the single bonded oxygens in the sulfate group attacks the carbon that the nitrogen group is bonded to, forcing the substitution. These types of reactions typically take place in high-NO_x environments, where many species with multiple N-containing groups can be found.^{49,50} These CHONS (organosulfates with nitrogen groups bound to them) are typically either primary or tertiary in nature. Primary CHONS have the sulfate group bound to a carbon that has only one carbon group attached to it whereas tertiary CHONS are bound to a carbon that have three other carbon groups attached. Earlier studies have shown that while both primary and tertiary CHONS are derived from SOA reactions of isoprene-derived epoxides, tertiary CHONS are typically formed through acid-dependent hydrolysis reactions.⁴⁹ Also, primary CHONS are generally fairly stable against nucleophilic substitution reactions whereas tertiary OSs undergo hydrolysis to form polyols and sulfuric acid.⁴⁹ An example of this mechanism can be seen below in Figure 4.

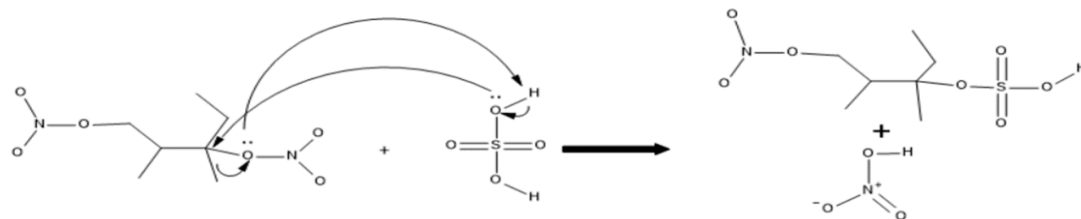


Figure 4–Substitution reaction of an organic nitrate group by sulfate

OSs can also form through the heterogeneous oxidation of unsaturated compounds. This mechanism involves sulfate anion radicals, which break a double bond in the structure allowing the sulfate group to attach.^{31,51} Sulfate radicals are formed in the atmosphere when sulfate compounds are oxidized in the presence of transition metals

such as manganese, or by reacting with OH radicals.⁵¹ Figure 5 demonstrates how these radicals break double bonds and attach to the structures.

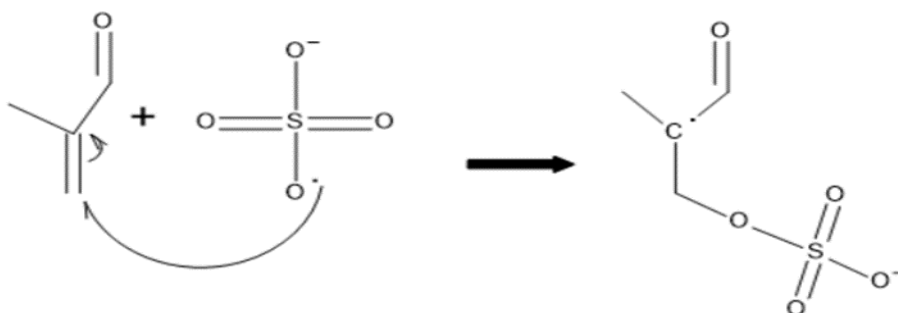


Figure 5 –Sulfate Radical breaking a double bond

It should be noted that OH radicals can also lead to the formation of OSs through a radical-radical reaction. In these reactions one OH radical pulls an electron from a compound, another OH radical pulls an electron from sulfuric acid in the atmosphere, and the two react to form an OS.⁵²

Another mechanism by which OSs can form is through esterification. This particular mechanism has been proposed as an explanation for how glycolaldehyde reacts with atmospheric sulfur.⁵² Glycolaldehyde is a common VOC found in the atmosphere and is produced from ethane, isoprene, and biomass burning. These types of reactions have been observed in experiments with very low levels of NO_x .⁴⁰ This is likely due to the fact that when NO_x reaches higher levels other organic acids compete with sulfuric acid or sulfate for the esterification of alcohols.³⁴ An example of the esterification reaction can be seen in Figure 6.

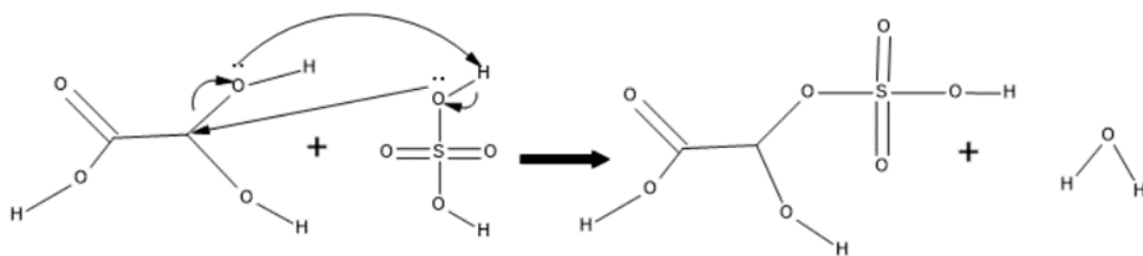


Figure 6 –An example of the esterification process leading to OS formation

More recently, OSs and sulfonates derived from anthropogenic precursors, such as polycyclic aromatic hydrocarbons (PAHs) or alkanes ≥ 10 carbons, have been observed in both laboratory-generated and field aerosols.^{10,42,43} A large presence of alkane-derived OSs, denoted by low double bond equivalence (DBE), have also been recently revealed in polluted urban areas.^{10,43,53} Compounds with low DBE values will typically only have one or zero double bonds in their chemical structure.⁴³ These new classes of OSs indicate that anthropogenic precursors may contribute significantly to the total organic sulfur within fine particulate matter (PM_{2.5}).^{43,53}

The total concentration that biogenic and anthropogenic sources contribute to the total concentration of OS in the atmosphere varies throughout the year. Changes in weather conditions can also impact the concentration of OS present in the atmosphere.^{2,15,48}

The purpose of this research is to classify, determine origin, and analyze seasonal variations of PM_{2.5} OS derived from both biogenic and anthropogenic sources from filters that were collected from August 2012 to May 2013 in Towson, Maryland. This longer sampling duration is rare in comparison to similar studies that have looked to categorize OSs in the past.^{12,48,54} Because of this, the longer sampling duration provides a unique

opportunity to examine the seasonal trends of OS species, as well as any possible correlations between OS and weather conditions.

II. Methodology

IIA. Preliminary Work

Sampling Site and Weather Data

The sampling site for this experiment, shown in Figure 7, is located in the Mid-Atlantic region of the United States in Towson, Maryland. The mid-Atlantic region of the U.S. can be classified to have a polluted atmosphere. This is due to non-attainment of certain criteria pollutant standards such as ozone and fine particulate matter. This is especially true in summer months.^{55,56} The local air mass is influenced by several large urban centers. This includes Washington D.C. and Baltimore to the south, and Philadelphia and Pittsburgh to the north. The location of the sampler was approximately 43 m above ground level on the roof of the Towson University Glenn A Residence Tower. The Towson University campus is located on York Road, which is a major north/south thoroughfare that connects Baltimore, MD, 11.25 kilometers to the south and York PA to the north. The campus is just to the south of the 695 beltway and to the northeast of Interstate 95. Emissions from vehicles peak during the morning and afternoon rush hours.⁵⁷ In order to correlate weather data to OS concentrations meteorological data were collected from the KMDTOWSO2 weather station, located 1.6 km south of Towson University.

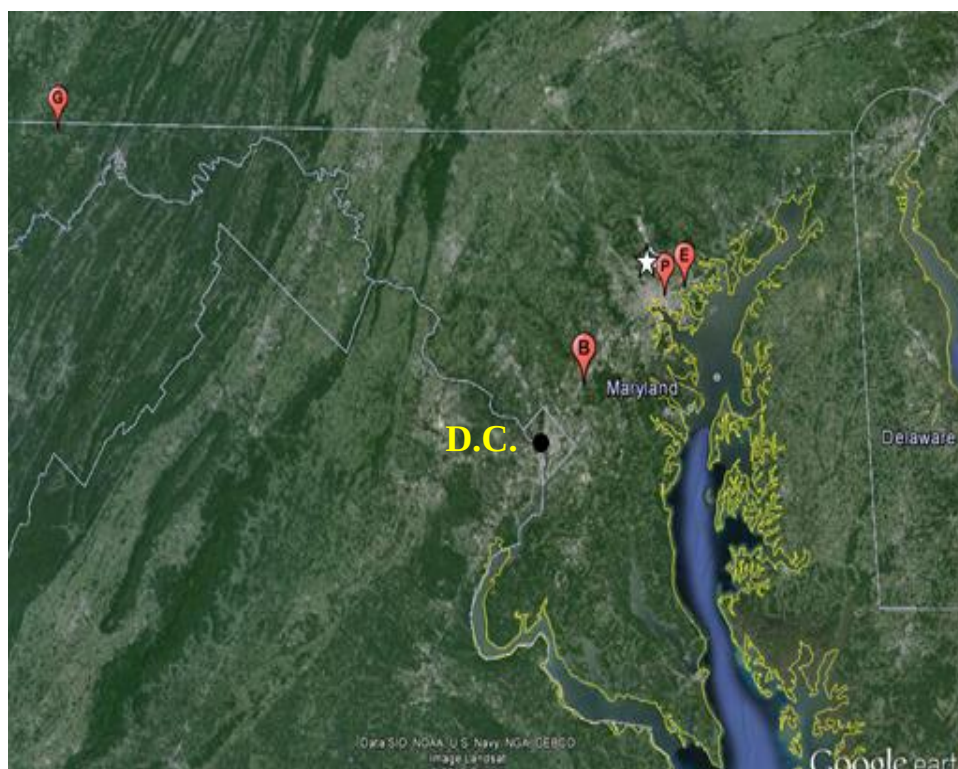


Figure 7- a) Map of sampling site. Star indicates location of Towson University Sampling site. Other sampling locations include the Ponca St super sampling site denoted P, and EPA sampling locations at Essex (E), Grantsville (G), and Beltsville (B). Maps prepared using Google earth.

Prior to the start of this study, several sampling campaigns have been performed in this region. For example, Discover-AQ provided detailed information that related to the levels of O₃, SO₂, NO₂, CO, and VOCs in this region during July 2011.^{56,58,59} Airborne measurements in summer months were collected from 1997-2003 as part of the Regional Atmospheric Measurement, Modeling, and Prediction Program.⁶⁰ In 2002 a supersite was established in the industrial region of eastern Baltimore. This industrial area includes roadways and tunnel systems that eventually converge near the Port of Baltimore. The purpose of this supersite was to measure pollution events from February to November 2002.^{26,61-63} These studies established a link between the concentration of

local sulfate aerosol and transport that was occurring from both local and regional coal-fired plants.^{62,64} These studies have helped to determine the sources and meteorological effects on aerosols in the Mid-Atlantic region.²⁶ However, these studies focused more on broad classes of gas- and aerosol-phase pollutants than organic aerosol composition. They also took place on relatively short time scales.

Weather data were collected for temperature, precipitation, ultraviolet (UV) radiation, wind speed and direction, and relative humidity (RH) at 5 minute intervals. Other parameters, including trace gas concentrations, PM_{2.5} loadings, and sulfate concentrations in PM_{2.5} were obtained from three local EPA monitoring stations. The stations are located in Essex, MD, to the southeast of the sampling site (39.31 °N, 76.47 °W), Grantsville, MD to the west of the sampling site (39.71 °N, 79.01 °W) and Beltsville, MD to the southwest of the sampling site (39.06 °N, 76.88 °W). These stations were chosen because of their relative location to the sampling site. The measurements taken from Grantsville are likely to represent atmospheric transport from the west. This air mass passes over the northern Ohio River Valley, and thus would likely have a higher concentration of OS. Beltsville measurements are representative of transport from the southwest and the Washington, D.C. area. Finally, Essex measurements represent air mass coming in from the east, and would likely have the lowest concentrations of OS as any air mass from this direction is coming primarily from the Chesapeake Bay and the Atlantic Ocean.

Precipitation rarely occurred on sampling days and when it did precipitation amounts were typically less than 1.5 cm. This is because rainy days were specifically avoided as sampling days due to the fact that excess precipitation typically results in

samples with little OS present. OS and other SOAs have a tendency to stick to water molecules, and excessive rainfall can lead to a “wash out”. Relative humidity (RH) for the sampling days ranged from 75-91%. Figure 8 shows the average temperature, relative humidity, and precipitation at the sampling site. Measurements were recorded at 5 minute intervals from the KMDTOWSO2 weather station located 1.6 km south of Towson University campus.

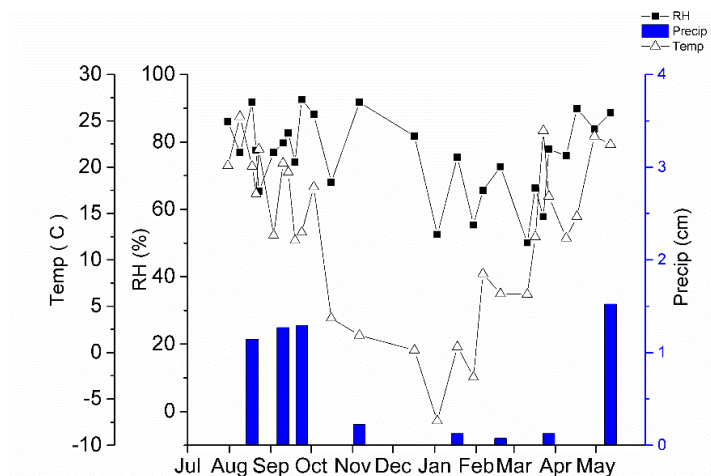


Figure 8-The temperature, relative humidity, and precipitation levels measured at the sampling location.

Trace gases (O_3 , SO_2 , NO_2 , and CO) and mass loadings ($PM_{2.5}$) for the sampling period were also recorded for correlation and analysis purposes from the three local EPA monitoring stations located in Grantsville, Essex, and Beltsville, MD. The average measurements for these parameters at these sites can be seen below in Table 1.

Table1: Average EPA Measurements for Trace Gases and Mass Loadings					
	O_3 (ppb)	SO_2 (ppb)	NO_2 (ppb)	CO (ppm)	$PM_{2.5}$ (mg/m³)
Grantsville	35.08±9.53	0.97±0.91	N/A	0.17±0.03	7.73±3.25
Beltsville	25.28±10.67	0.54±0.49	8.92±5.15	0.24±0.08	10.20±4.76
Essex	24.25±10.95	1.43±0.81	11.82±6.04	0.24±0.14	9.76±5.98

The observed sulfate concentrations are similar to the concentrations observed at the Ponca St. Supersite during non-summer months.⁶¹ Beltsville and Grantsville sulfate concentrations are well correlated ($r = 0.81$), and sulfate from Beltsville and Essex are moderately correlated as well ($r = 0.42$). The sulfate concentrations between the Essex site and Grantsville site show no correlation.

Filter Sampling and Collection:

Thirty-two filters were collected using a Tisch sampler with a flow rate of approximately $1 \text{ m}^3/\text{min}$ between September 2012 and May 2013 in order to assess the chemical composition of local SOA. In order to preserve the filters, they were stored in a -80°C freezer. The filters were moved to a -5°C freezer in the fall of 2015 due to a technical issue with the -80°C freezer.

Solvent Study

Before analysis was started a solvent study was conducted in order to determine the best solvent to conduct the filter extractions. The solvent study was necessary because recent studies have suggested that less polar solvents, such as an acetonitrile (ACN)/toluene mixture, are better suited to extract alkane derived OSs.^{42,43} However, many earlier studies used either a polar solvent (methanol) or a mixture of a polar and non-polar solvent.^{34,48,65} Despite this, no one has conducted a study to determine which solvent is the most effective for this type of analysis.

Multiple samples were taken from May-July of 2015 and were extracted with different solvents with different degrees of polarity. Solvents included methanol (Sigma-Aldrich, Chromosolv® grade), dichloromethane (DCM) (Sigma-Aldrich, Chromosolv® grade), and a 50/50 mixture of the two. Although not historically used as a solvent for

OSs, DCM was chosen based on the results of extracts done with this solvent in earlier field studies.^{19,24} Methanol has a dielectric constant of 32.7, whereas DCM has a dielectric constant of 8.93. This means that methanol is more relatively polar than DCM. Therefore, by examining the relative concentration of compounds that each solvent was able to extract from the filters, it was possible to determine if using a solvent that was a mixture of a relatively polar and nonpolar compound (methanol and dcm) was more effective than simply using the relatively polar solvent (methanol) alone.

To compare the solvents, punches were taken from the same filter sample. Each filter was cut using a 47 mm diameter hole punch and covered with 15 mL of the appropriate solvent. Samples were then sonicated in an ice bath for 45 minutes, extracted, and analyzed using both ultra-performance liquid chromatography/electrospray ionization-high resolution mass spectrometry (UPLC/ESI-MS) and gas chromatography/electron impact- mass spectrometry (GC/EI-MS) analysis. Methodology for the operation of these instruments was adopted from previous studies.^{10,12,29,34} By analyzing the resulting spectra, it was possible to qualitatively determine which solvent was able to extract the greatest quantity of compounds since both solvents were used on the same filter. This made it possible to ultimately decide which solvent would be best suited for the experiment. It should be noted that while GC/EI-MS analysis was not used for OS it still needed to be done for the solvent study portion of this experiment because other future work with these filter samples will need to use GC/EI-MS analysis.

The results of the UPLC solvent extraction can be seen in Figure 9 and the GC/MS extraction can be seen in Figure 10. In both cases the 50/50 mixture is compared to methanol alone.

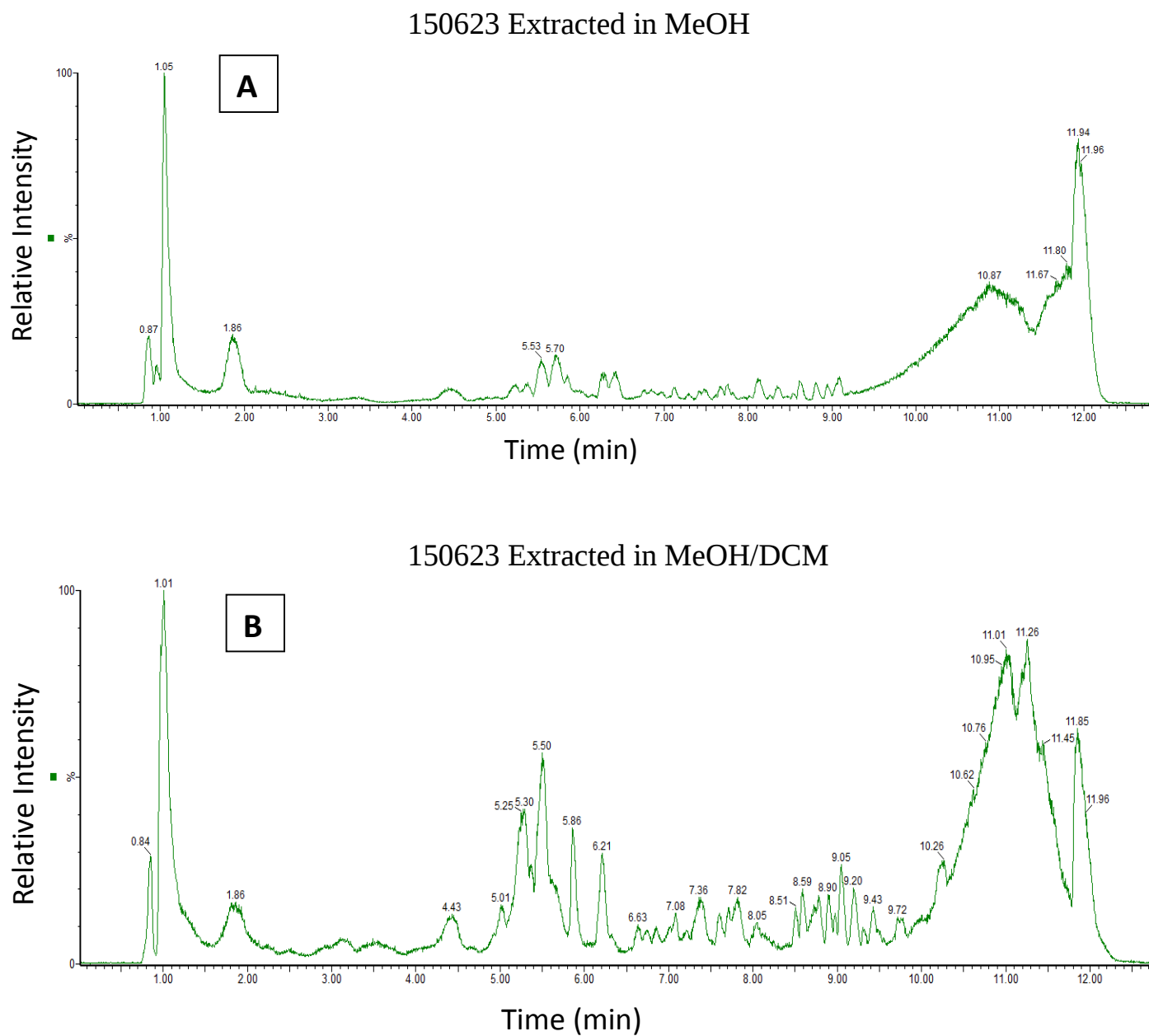


Figure 9-UPLC BPC performed in methanol (top) compared to UPLC extraction performed in a MeOH/DCM mixture.

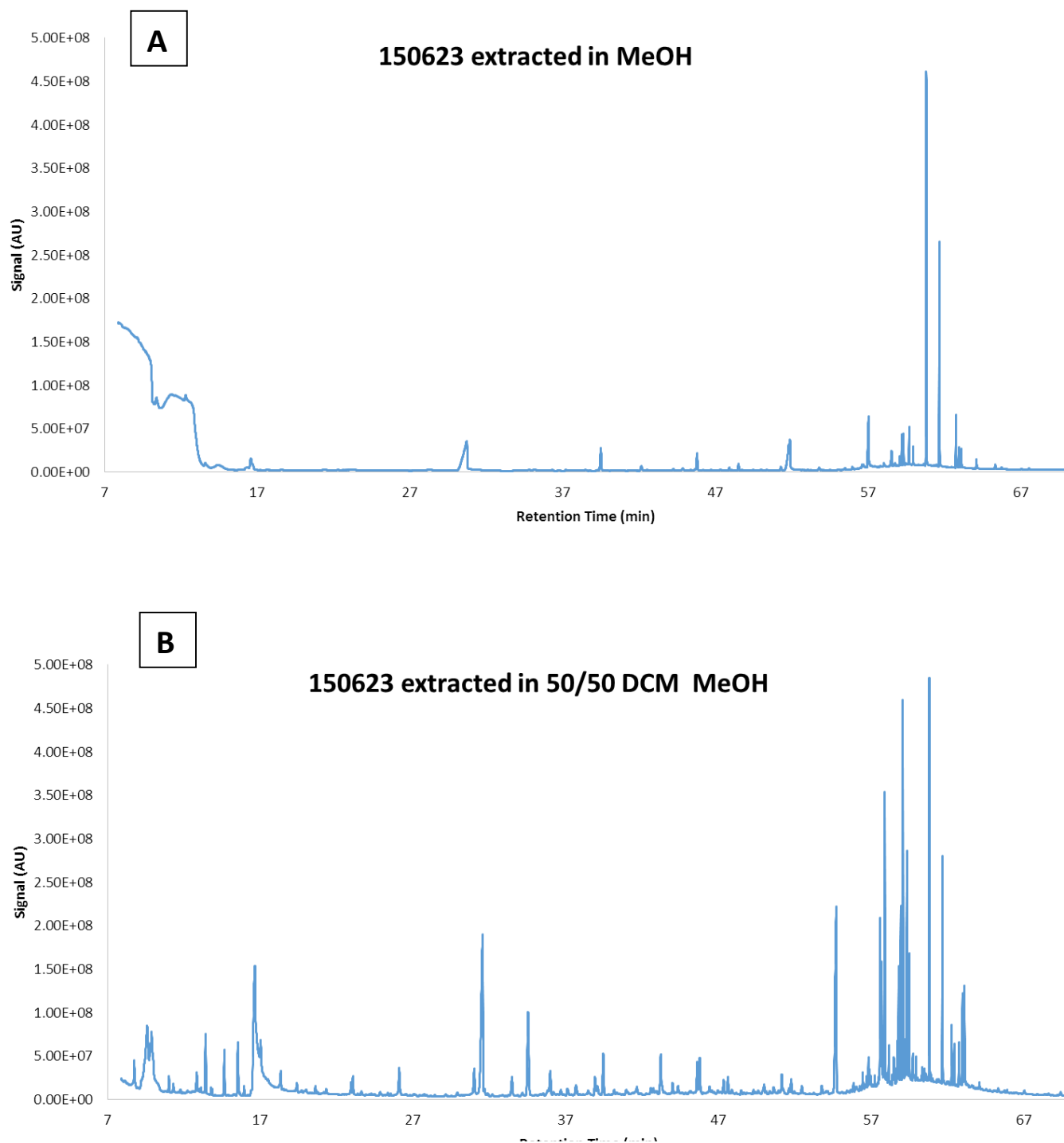


Figure 10- GC/MS BPC done in Methanol (top) Compared to UPLC extraction done in a MeOH/DCM mixture

After examining the results of the UPLC/ESI-MS and the GC/EI-MS it was qualitatively determined that the best solvent to use for extraction purposes was the 50/50 mixture of methanol and dichloromethane. Since OS and SOAs can be both polar and non-polar in nature, the best extraction solvent would likely be one that has both polar and non-polar qualities. Every extraction that was conducted shows a visibly discernable difference in the mass spectra results between the three solvents (methanol, DCM, and a 50/50 mixture of methanol and DCM) When the samples were extracted with the 50/50 mixture the relative intensity of the spectrum peaks were greater, spectra peaks were sharper, and there were more compounds found when mass extraction was done. This is true for both UPLC and GC analyses. This means that by extracting the samples in a 50/50 mixture it is possible to extract more OS from the filter samples.

IIB. Sample Filter Extraction and UPLC/ESI-MS:

UPLC/ESI-MS was used to analyze the filter samples for the presence of OSs. This is because OSs cannot be detected through other quantitation methods, such as GC/EI-MS. The ionization method is softer in UPLC/ESI-MS compared to GC/EI-MS.^{33,42} This means that the UPLC ionization causes less fragmentation and more easily allows analysis of the parent ion of the compounds. It also made more sense to use UPLC for OS analysis as sulfates are typically less volatile than some other secondary organic aerosols.^{33,42} Finally, by combining the UPLC/ESI with mass spectrometry/mass spectrometry analysis, it was possible to look for specific peaks in the mass spectra that are indicative of OSs. To generate the samples, filters were cut into quarters and each sample was spiked with 815 μL of 12.24ppm ketopinic acid as a recovery standard. Using a pair of sterilized tweezers, the samples were folded and inserted into an extraction vial

that had been previously rinsed with HPLC grade methanol. The filters were sonicated in 15 mL of 50/50 HPLC-grade methanol and dichloromethane for 45 minutes. An ice bath was used in order to prevent the water in the sonicator from heating up and compromising the sample, since the heat could have caused the organic compounds to volatilize. After sonication was complete, each of the extracted filters was transferred to a new vial labeled for storage. The remaining contents of the extraction vials were transferred to a new set of pre cleaned vials using a syringe and 0.2 μ m hydrophilic filter. Samples were dried with nitrogen at a flow rate of 3 L/min. After the dry-down process, samples were reconstituted in 150 μ L of a 50/50 mixture of methanol and dichloromethane. Samples were spiked with 20 μ L of a mix of 1.1 ppb camphor-10-sulfonic acid (m/z 231) and 1.02 ppb salicylic acid as internal standard. The samples were analyzed using a C18 HSS₃, 2.1mm x 100mm, 1.7 μ m column and the flow rate was set to 0.3 mL/min. The UPLC instrument was operated in negative mode, as earlier studies have shown that operating in this mode provides the best sensitivity for the detection of OSs.^{34,66} This holds especially true for OSs that were formed in highly acidic environments, such as sulfate esters. A more detailed explanation of the methodology can be seen in several other studies.^{29,34}

When examining mass spectra results for OSs, the first thing to look for are m/z values of 96.95, 95.95, or 79.95. These m/z values correspond to the presence of (HSO_4^-), (SO_4^{2-}) and ($\text{SO}_3^{\bullet-}$) respectively as these are common fragments from OS compounds. The presence of these ions, especially when they have a high relative abundance, strongly suggests that the observed species is an OS.^{34,66} In order to determine the type of OS present on the filters a list was compiled from multiple prior

studies that had detected different types of OS.^{29,33,43,66} These OS were both biogenic and anthropogenic in nature. The OS species on the master list were then extracted from the extracted ion chromatogram in order to determine what was present on our sample filters. This was done for each filter, resulting in more than 7,500 individual values for our analysis. By extracting the 96.95, 95.95, and 79.95 ions using MS/MS analysis, it was possible to determine if there were any other OS species in the filters that had not been seen in other studies. Since these ions indicate the presence of OS, using the MS/MS spectra to analyze the formula and retention times of any parent mass with these ions made it possible to determine if any observed OS were previously unidentified. By compiling such a large data set it was possible to correlate different types of OS to each other. It was also possible to correlate the observed OS species to varying weather conditions and changes in the seasons.

IIC. Weather Data and the HYSPLIT Model

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, weather information from the National Oceanic and Atmospheric Administration (NOAA), and weather data obtained from the National Weather Service station in Towson, Maryland were used in order to understand the link between chemical composition and chemical sources in regards to changing chemical transport. Historical meteorological data were also used to examine any potential correlation between various weather conditions such as humidity, wind speed, and wind direction to the amount of deposition observed on the filters. This provided better insights as to the aerosols' origin. These data sets, as well as access to the HYSPLIT model, are available online.

Meteorological data were used to examine backward trajectory of the air mass

from which our samples were taken. The HYSPLIT model provides insight into how the sampled air mass arrived at the sampling location. Through the use of backward trajectory modeling it was possible to examine potential sources of the sampled OSs, the velocity of the air mass, how the air mass moved in the air column, changes in the boundary layer, and changes in trajectory over the sampling duration. Samples were ranked from greatest total OS concentration to lowest total concentration and compared to the HYSPLIT trajectories in order to examine any qualitative impacts that these trajectories had on the observed OS. The exact latitude and longitude of Glenn Tower A (39.392 °N, 76.610 °W) was used as the starting point. For each modeled sample, the HYSPLIT model was run twice. The first run was performed at three heights; 500, 1000, and 1500 meters. These heights represent the bottom, middle, and top of the atmospheric boundary layer (ABL) where the majority of OSs transport occurs.⁶⁷ The ABL is the lowest portion of the Earth's atmosphere, and expands from near ground level to just below the elevation where cumulus clouds form. Its behavior is directly influenced by contact with the planetary surface.⁶⁷ Any transport that is occurring below the boundary layer is likely due to turbulence at the ground level, and doesn't provide an accurate depiction of source. Any air mass above this level is considered the "free atmosphere" and is not being affected by the surface of the earth.⁶⁷ By selecting these three elevations we were able to examine the potential origin of the aerosols as well as how they moved in the ABL. The backward trajectory runtime started when the sample was collected and ran for 96 hours. This resulted in a model-generated trajectory that made it possible to examine where the air mass was likely coming from and how much mixing was occurring in the air column itself. The model was also run a second time at the 500 meter mark at

multiple time steps. This backward trajectory was set for 24 hours, with a new trajectory starting every 4 hours. This generated 6 local trajectories which made it possible to examine any changes in movement as the air mass approached the sampler. It was necessary to do these two runs separately because the HYSPLIT model cannot simultaneously perform a time step trajectory and a multiple height trajectory.

IID. Determining Correlation

Having a large data set from nearly a year of sampling made it possible to examine correlations between different observed OSs, as well as correlations between OSs and weather conditions. This sort of correlation analysis would have been much more difficult if the sampling duration had been limited to only a few months. After determining which OSs were derived from biogenic sources and which were derived from anthropogenic sources a table of values was made that included all mass values as well as values for weather conditions. These values were correlated to each other and the correlation coefficient was determined to represent the strength of the correlation between these values. Using Pearson Product-moment correlation coefficient values, it was determined that, for the number of samples collected in this study, a Pearson's correlation coefficient (r) of 0.4 represents correlation at the 95% level. R values of 0.5 represent correlation at the 99% level. Table 2 shows a color key used to differentiate the level of correlation between two values. If a value has no color coding, the r -value was less than 0.4 and showed a correlation under the 95% level. The full table of all correlations can be seen in Appendix 1.

Table 2-Correlation Table Key

Possitive r correlation (95%)	
Possitive r correlation (99%)	
Negative r correlation (95%)	
Negative r correlation (99%)	

III. Validations

Due to the nature of the samples, the compounds on the filters are prone to artifacts. This means that the analysis and preparation of the samples can result in the presence of compounds that would not be observed otherwise.¹⁵ This can occur when the samples are being analyzed, if they are exposed to light for an extended period of time, and when they are not stored at the appropriate temperature.¹⁵ In order to correct for these artifacts, two types of blanks were utilized; field blanks and method blanks. Field blanks are filters that were put on the sampler but the sampler was never turned on. This provides a way to correct for any compounds that are found on the filters as a result of the filter being placed outside on the sampler. Method blank filters are filters that were pre-baked and spiked, but nothing else had been done to them. By using method blank filters it was possible to correct for any contamination that exists on the filters themselves. Solvent blanks were analyzed to ensure that there is no contamination in the solvents or instrumentation themselves.

After the UPLC/ESI-MS analysis was performed for each sample, masses that indicate OS presence were selected based on the finding of earlier studies were extracted and the program used in the UPLS/ESI-MS generated a total area value that each extracted mass had in each sample.^{10,29,31,34,42,51,66,68} These raw area values were then converted to values in ng/m³. To do this, each OS was assigned a surrogate compound.

These surrogates were analyzed using the UPLC/ESI at five different concentrations (0.1, 1, 5, 10, and 50 ppm) in order to generate a calibration curve. Using this calibration curve, it was possible to convert the samples from raw areas to a concentration in solution. At this point, it was necessary to multiply some of the values by a dilution factor, as some of the samples had needed to be diluted before analysis in order to prevent damage to the instrument. Sample concentration in solution was then converted to ng/m^3 and multiplied by the volume of air sampled in order to determine the concentration in the air using the following formula:

$$\frac{\text{Concentration in solution (mg/L)} * \text{Volume of solution (17} \times 10^{-6} \text{ L)} * 1 \times 10^6 \text{ ng/mg}}{\text{Volume of air sampled (m}^3\text{)}}$$

The surrogates used for these conversions can be seen in Table 3. Surrogates were chosen based on the retention time of the compound, with each compound being matched to the surrogate with the closest retention time. If a compound closely matched two retention times the surrogate with the more similar formula was chosen to represent the compound. These surrogates have also been used in previous studies for similar types of analysis.^{29,33,40,42}

Table 3-Standards used as surrogates for observed OS species

Name	Formula	m/z	Retention time (min)
Glycolic Sulfate	$\text{C}_2\text{H}_3\text{O}_6\text{S}^-$	154.96	1.78
Lactic Sulfate	$\text{C}_3\text{H}_5\text{O}_6\text{S}^-$	168.98	1.83
Propyl Sulfate	$\text{C}_3\text{H}_7\text{O}_4\text{S}^-$	139.01	3.27
Octyl Sulfate	$\text{C}_8\text{H}_{17}\text{O}_4\text{S}^-$	209.08	11.53

It should be noted that while internal standards were added to each of the samples, the staff at UNC did not analyze the recovery standards on the UPLC. Ketopinic acid was

added as a recovery standard but was not detected in the UPLC analysis. GC/MS analysis has shown an 85% recovery of ketopinic acid when it is used as a recovery standard.

IV. **Results/Discussion:**

IVA. UPLC/ESI-MS:

A full table of all biogenic and anthropogenic compounds that were found on the filter samples can be seen in Appendix 2. This table includes the suggested formula, mass weight, and average concentration in ng/m^3 , precursor group, and retention time of each observed compound. The most prevalent OSs originating from biogenic sources originate from either isoprene, isoprene derivatives such as methyl vinyl ketone and methacrolein, or monoterpenes. Due to limitations of sampling few samples were taken in the months of August and June. As such, the values for these months are the values observed for those individual dates.

The OS observed in the UPLC/ESI-MS spectra results were characterized as being either derived from biogenic sources or derived from anthropogenic sources. It was possible to determine which OS belonged to which category because prior research lists common formula and m/z values for OS that we observed and have determined the nature of their origin.^{31,33,42,48,51,68-70} The observed biogenic compounds are emitted by plants as they photosynthesize in summer months and are a common source of biogenically emitted VOCs. OS that originate from anthropogenic sources typically form from reacting with various PAHs and long-chain alkanes.³¹ After OS was characterized as either biogenic or anthropogenic it was possible to examine the changes in total concentration of these OS and correlate them to changes in the seasons.

Biogenic OS

Isoprene was the most prevalent biogenic precursor to the observed OS shown in Appendix 2, with the greatest concentration originating from m/z 215.02 ($C_5H_{11}O_7S^-$). Three isomers of this compound were observed, and two of them were observed consistently throughout the samples. In one instance the concentration of this m/z 215.02 surpassed 50 ng/m^3 . During the autumn months of our study the concentration of this compound averaged $3.57 \pm 0.7\text{ ng/m}^3$. Measurements were highest in September and dropped as the weather got cooler. The three isomers of m/z 213.01 ($C_5H_9O_7S^-$) also had relatively high values, with spring averages of one isomer reaching $3.25 \pm 6.5\text{ ng/m}^3$ during the spring. In August the two isomers of mass $C_4H_7O_7S^-$ reached concentrations of 3.16 ng/m^3 and 1.27 ng/m^3 , respectively. Other OS that are biogenic in nature can be seen in the samples but have a much smaller concentration, typically averaging between $0.1\text{--}2\text{ ng/m}^3$. This includes compounds derived from the oxidation of terpenes. Six of the OS that were identified (OS-223.03, -239.02, -251.06, -267.05, -279.05 and -283.05) are terpene-derived and contribute to a total average of $4.5 \pm 0.4\text{ ng/m}^3$. The sum of all OS totals an average of $120 \pm 60\text{ ng/m}^3$, and the trends of some of the most prevalent biogenic OS species can be seen in Figure 11.

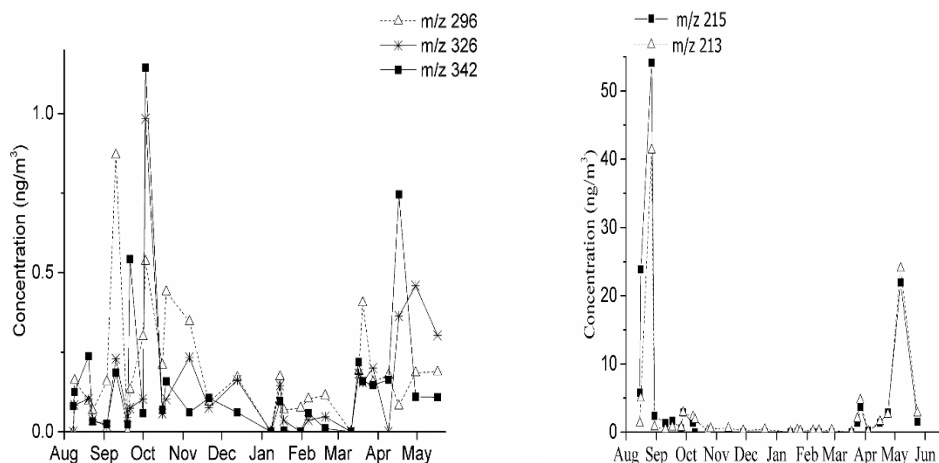


Figure 11 –An example of observed biogenic trends throughout the sampling duration.

These results are comparable to OS concentrations seen in several locations in Asia^{12,71} and Europe⁴⁷. In Asia, Ma et al observed biogenic OS ranging from 0.5-26.1 ng/m³ depending on the sampling day, and isoprene derived OS dominated.⁷¹ Stone et al reported total OS concentrations ranging from 100-2000 ng/m³, depending on location.¹² In Europe, Nguyen et al reported average OS concentrations ranging from 0.2-7.4 ng/m³, depending on the species.⁷²

Regions with larger concentrations of biogenic OS are typically more prevalent in more rural regions, such as in southeast USA.^{73,74} Isoprene and compounds derived from isoprene have been shown in previous studies to significantly contribute to PM_{2.5} in the mid-Atlantic region.⁴⁵ In some cases isoprene concentrations in SOA air mass ranged from 5ng/m³ to 50ng/m³.^{45,58} Isoprene-derived VOCs are, in fact, the most abundant VOCs in the mid-Atlantic.^{58,75} The isomers of m/z 215.02 have been observed in many locations around the world in both rural^{29,34} and urban areas.^{76,77} The three isomers of m/z 213.01 (C₅H₉O₇S⁻) have been observed in previous studies as well, and are formed

from the photooxidation of isoprene in low-NO_x conditions.³⁴ Conversely, the isomers observed at *m/z* 198.99 are formed when isoprene is oxidized in high-NO_x conditions.^{29,40} The prevalence and relative abundance of both of *m/z* 213.01 and *m/z* 198.99 suggests that the OSs forming from biogenic sources in this region are doing so in regions with relatively high NO_x conditions and relatively low NO_x conditions.

Anthropogenic OS

Anthropogenic OS in the samples generally have a high *m/z*, and in our study, C₇ and C₉ species are the main contributors. The observed OS also have low DBE values; typically between 0-1. Some of the observed anthropogenic masses seen in Appendix 2 include *m/z* 207.07, 209.05, 211.06, and 237.08. These compounds have observed in an earlier study where they were determined to be alkyl-derived OS.⁵³ On average, the concentration of each of these OSs accounts for 0.1-0.4 ng/m³ of the total 120 ± 60 ng/m³ OS collected in the samples. The parent ion observed at *m/z* 279.13 (C₁₂H₂₃O₅S⁻) was also recently identified in a chamber study from the oxidation of dodecane and contributes an additional 0.3 ng/m³ to the observed SOA total.⁴² An example of the trends observed from the anthropogenic OSs can be seen in Figure 12.

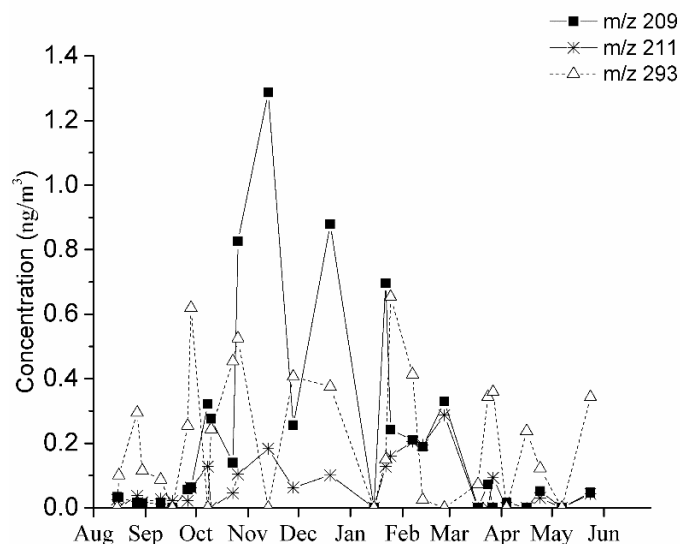


Figure 12—An example of observed anthropogenic trends throughout the sampling duration.

The formation of OS from anthropogenic emissions varies depending on the location. In the mid-Atlantic region of the United States, the observed levels of these OS are generally higher in close proximity to urban centers.^{58,75,78} As such, it is not unreasonable to think that there would be an increased concentration of this type of OS in air sampled from the Towson region. Anthropogenic OS can form when PAHs and long chain alkanes are oxidized in the atmosphere.³¹ Compounds with long chain alkanes and PAHs can be identified because they have low DBE values.⁴³ The presence of long-chain alkanes with low DBE values provides credibility to the conclusion that these observed OS were anthropogenic in nature.^{43,79}

Anthropogenic OS like the ones observed here have only recently been observed in ambient aerosols, and have been gaining increasing attention in more recent literature.^{16,33} This is because historically only biogenic OSs were analyzed, and very little research has been done in regards to anthropogenic OS.^{10,42,43,53,71,80} Interestingly, some of these more recent studies have suggested that some anthropogenic OS act as

surfactants.⁴³ Surfactants are compounds that lower surface tension, either between two liquids or between a liquid and a solid. This means that these OS have the potential to alter SOA hygroscopic and radiative properties.^{43,81}

Biogenic and Anthropogenic OS Correlations

Most of the OS derived from biogenic sources correlated strongly with other biogenically derived OSs. For example, many of the OS derived from isoprene strongly correlated with the OS derived from monoterpenes. This is especially true for OS derived from α -pinene. Of the 54 observed OS species, 20 of the biogenic OSs correlate at the 95% level and 31 correlate at the 99% level to other biogenic OS species.

Anthropogenic species show some level of correlation between other anthropogenic species, but to a lesser extent that is observed between biogenic species. Two compounds derived from limonene (m/z 239.02 and 251.06) correlate at the 95% level with anthropogenic alkyl-derived OSs. In total, 9 of the observed anthropogenic OSs correlate at the 95% level and 12 correlate at the 99% level to other OSs that are potentially derived from anthropogenic sources. In addition, a total of 9 biogenic OSs correlate at the 95% level to anthropogenic OSs, and 8 biogenic OSs correlate at the 99% level to anthropogenic OSs. An example of the correlations between different OS species can be seen in Table 4.

Table 4-Example of OS Correlations to other OS Species

		C6H13O4S-	C4H7O6S-	C5H7O6S	C5H9O6S-	C4H7O7S-
		alkyl OS	alkyl OS; Isoprene	Isoprene	alkyl OS, isoprene	isoprene
Suggested Formula Precursor		181.054	182.99688	194.9963	197.01253	198.991252
C5H9O7S-	isoprene	0.508160788	0.735804904	0.59539262	0.680035725	0.62915733
C5H9O7S-	isoprene	0.521803749	0.615670874	0.41188396	0.585061792	0.715964
C5H9O7S-	isoprene	0.429371721	0.414651651	0.03129509	0.267090431	0.65812805
C5H11O7S-	isoprene	-0.04810165	0.781807198	0.68926135	0.603297108	0.47023879
C5H11O7S-	isoprene	0.204499161	0.6259399	0.1740962	0.396842696	0.83861247

The observed CHONS compounds correlate well with terpene and isoprene-derived OSs. 5 of the 9 identified CHONS in Appendix 2 are also assigned to monoterpene chemistry (OS -294.06, -296.04, -326.05, -342.05 and -373.06), and could represent a major source of CHONS compounds identified in our study, though the CHONS are minor contributors ($\sim 1.6 \text{ ng/m}^3$) to the overall OS mass. The trends of some of the most prevalent CHONS species can be seen in Figure 13.

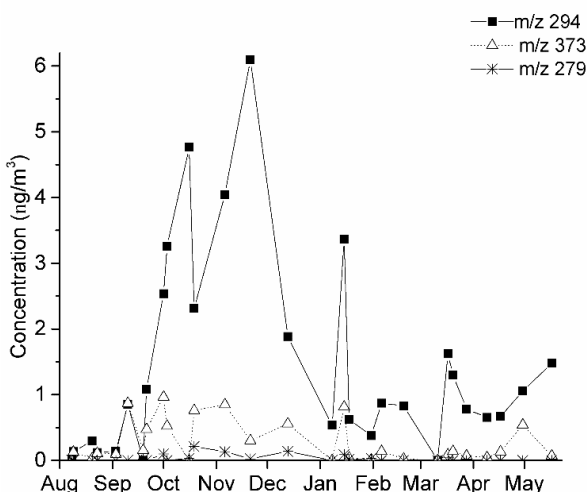


Figure 13 –Observed CHONS trends throughout the sampling duration.

The strong correlation between observed biogenic OS species indicates that many of these biogenic OSs are being emitted into the atmosphere at relatively the same time, and from similar sources. The strength of these correlations is also likely due to the fact that many of the observed OS species in Appendix 2 are derived from isoprene.

It is also important to mention that Riva et al. (2016) recently reported the formation of isobaric OSs (OS-267.02, -279.05, -297.06 and -326.05) from the gas-phase oxidation of long-chain alkenes, which were previously identified as monoterpene-derived OSs.³⁷ This means that CHONS could potentially be derived from both biogenic

and anthropogenic sources, even though they have historically only been classified as biogenic.²⁰ Therefore the CHONS OSs identified in this study might arise from different sources, especially during colder months.

Variations and Correlations between Seasons

The OSs derived from isoprene observed in Appendix 2 are very prevalent during the warmer sampling days and in the warmer parts of the year. These biogenic OS correlate strongly with temperature values ($r > 0.65$) and concentrations observed on warmer sampling days are an order of magnitude greater than concentrations observed in the late fall and winter months. As the seasons change, these OS begin to decrease until they all but disappear in the later fall months, only to reappear in late March. Trends for other observed biogenic species of OSs are variable depending on the species. Some of the OS derived from terpenes peak during warmer months, exhibiting r values ranging from 0.42-0.53. Some of the masses related to terpenes (m/z 267.02 and 283.05) do not correlate with changes in the season or temperature. CHONS have more variable trends in regards to seasonal changes⁷¹. For instance, m/z 342.05 has a positive correlation with temperature and peaks in the spring. Other compounds with nitrogen groups observed in Appendix 2 have very little or no seasonal trends and stay at a consistent concentration throughout the year.

Unlike the OSs derived from biogenic sources, species derived from observed anthropogenic sources more frequently peak in the winter months. These anthropogenic OSs (m/z 209.05, 211.06, and 293.18) have a very strong negative correlation with temperature with r values typically near -0.6. It should be noted that the parent ions

observed at m/z 294.06 ($C_{10}H_{16}NO_7S^-$), and 373.06 ($C_{10}H_{17}N_2O_{11}S^-$), were previously assigned as OSs that form from the oxidation of monoterpenes, but these species follow the trends of anthropogenically derived OS and peak during the fall and winter instead of the spring and summer.²⁹ An example of the correlations observed between temperatures and observed OS species can be seen in Table 5, and the entire set of correlations can be seen as a part of the table in Appendix 1.

Table 5-Example of OS Correlations to Temperature

	<i>C4H7O7S-</i>	<i>C4H7O7S-</i>	<i>C7H13O5S-</i>
	<i>isoprene</i>	<i>isoprene</i>	<i>alkyl OS</i>
	198.991252	198.991252	209.04892
temp C	0.51950204	0.4552905	-0.56485783
temp high C	0.49597553	0.456345	-0.62222268
temp low	0.50956663	0.41415861	-0.46588233

The decrease in concentration of biogenic OS as the seasons change follows the patterns of isoprene emissions from broadleaf trees observed in previous studies.¹³ The strong correlation between temperature and isoprene derived OS has been observed in an earlier study conducted in the southeast, where SOAs derived from isoprene are abundant in the spring and summer but are not present in the fall and winter months.⁷⁴ Previous studies have shown that monoterpene derived OSs do tend to correlate with temperature and seasonal changes, but this correlation is much smaller compared to the correlation of isoprene.⁷¹ The variability observed in the CHONS lends more credibility to the theory that CHONS are derived from both biogenic as well as anthropogenic sources, since if they were only derived from one source it would be more likely that they would have a more distinct peak in frequency.

One possible explanation for the strong negative correlation between decreasing temperatures and increasing concentrations of anthropogenic compounds is the

prevalence of wood-burning stoves in the region and in Pennsylvania.^{82,83} These stoves are commonly used for heat in the winter months and lead to an increase in biomass burning.⁸²⁻⁸⁴ Earlier studies have shown that biomass burning leads to the formation of long-chain OSs and may be a significant regional source of these compounds.⁸⁵ Fuel and gasoline combustion is another likely contributor of anthropogenic OS as the sampling site is located close to a large urban center.⁷⁵ Considering the urban setting of the sampling site, it is also likely that anthropogenic VOCs from the combustion of fuel and gasoline also likely contribute to the total concentration of anthropogenic OSs.⁷⁵ The fact that two of the observed OSs that were previously assigned as monoterpenes could suggest that both of these OSs might also be formed from the oxidation of long-chain alkanes or other anthropogenic VOCs as well as from the oxidation of monoterpene.

IVD. HYSPLIT:

The results of the HYSPLIT model clearly show that air mass trajectory has an impact in the concentration of OS present in the samples. Figure 14 A shows an example of a HYSPLIT trajectory from a sample with a high concentration of OS originating from the northwest as a result of the trajectory's close proximity to several larger urban centers. Typically, air trajectories for the samples came from the south and the southwest, or only crossed a major urban center for a brief time. This results in an intermediate level of OS.

The 24- hour back trajectories for the April 30th sample (Figure 14 B) shows a slow-moving air mass originating over the Chesapeake Bay that remains at higher elevations for the majority of the sampling time, and then descends rapidly just before sampling

occurs. This is particularly true of the light blue time step that passes directly over the Bay.

The trajectory demonstrated in Figure 15 shows the impact that vertical lifting can have on the concentration of OS observed in a sample. Figure 16 shows the potential impacts that extreme weather phenomena can have on observed OS.

These changes in concentration are due to changes in trajectory and differences of origin. Trajectories from the west show that $\text{PM}_{2.5}$ sulfate that was recorded from the EPA site at Greenbelt correlates well ($r > 0.97$) with the presence of alkyl- derived OS ($\text{C}_8\text{H}_{15}\text{O}_4\text{S}^-$, $\text{C}_7\text{H}_{13}\text{O}_5\text{S}^-$, $\text{C}_7\text{H}_{15}\text{O}_5\text{S}^-$). Trajectories from the south that pass over the Beltsville EPA site demonstrate some correlation ($r > 0.88$) between $\text{PM}_{2.5}$ sulfate and isoprene derived OS. This includes the $\text{C}_4\text{H}_7\text{O}_6\text{S}^-$, $\text{C}_5\text{H}_7\text{O}_6\text{S}$, $\text{C}_5\text{H}_9\text{O}_6\text{S}^-$, $\text{C}_4\text{H}_7\text{O}_7\text{S}^-$, $\text{C}_4\text{H}_7\text{O}_7\text{S}^-$ species.

NOAA HYSPLIT MODEL
Backward trajectories ending at 1000 UTC 06 Nov 12
NAM Meteorological Data

Source ★ at 39.39 N 76.61 W

NOAA HYSPLIT MODEL
Backward trajectories ending at 1000 UTC 07 Jun 13
NAM Meteorological Data

Source ★ at 39.39 N 76.61 W

Meters AGL

Meters AGL

Figure 16- HYSPLIT model showing the trajectory of the air mass moving with Tropical Storm Andrea.

Samples with elevated OS concentrations pass close to major urban centers, including the Ohio River Valley, Philadelphia, or Pittsburgh (Figure 14a). They can also occur when the air mass passes over regions where excessive coal burning is occurring.^{62,64} An increase in OS concentration can also be seen in samples that have slow moving trajectories. Conversely, samples with lower OS concentrations occur when the air mass does not pass over major urban centers, or moves over urban centers very early or very late in the 24-hour sampling period.

Low OS concentrations could also be observed in samples that had fast-moving, higher elevated trajectories. Often these trajectories flow over the Atlantic Ocean or the Chesapeake Bay (Figure 14b). This type of phenomenon has been seen in other studies that examine the effects that bay winds have on pollutant levels in Baltimore.^{86,87} These studies have shown that bay breezes can lift pollution up into the atmosphere, leaving the air at ground level relatively clean.^{59,86,87}

The correlation values between the trajectories and the EPA site data indicate a link between OSs found in the samples and trajectories originating from the south and the west. The maximum elevation that the air mass reached during the sampling time also impacts the quantity of OSs in the samples. Typically, if the maximum elevation at the time of sampling was relatively low and the air mass did not experience a great amount of uplift during the sampling duration a higher concentration of OS would be observed on the sample. For example, the sample taken on 11/06/12 (Figure 15) has very low OS concentrations even though it passes directly over a major urban center. This is likely due to the fact that the air mass experienced uplift and was forced to a higher elevation,

making it unable to pick up and transport any OS that would have been present at lower elevations.

It is also important to note the possibility that tropical cyclone events may bring OSs up from the gulf coast. This phenomenon would explain the OS levels present on the sample taken on 6/07/13 (Figure 16). This sample should have been relatively clean because the air mass was over the Atlantic Ocean for nearly the entire duration of the backward trajectory. The trajectory also did not pass over any major urban centers. Despite this, the OS concentrations for this sample were unexpectedly higher than anticipated based on the trajectory. After examining the historical weather data, it was revealed that the air mass for this sample likely originates from winds generated by Tropical Storm Andrea, which made land-fall 6/05/13. A plausible explanation is that the near-hurricane force winds resulted in the transport of OS originating from more urban regions along Gulf of Mexico along the Eastern seaboard until it reached Towson, MD.

V. Conclusions

OSs are found in ambient fine particulate matter and serve as markers for different chemical process that result in the formation of SOAs. These compounds can substantially impact global climate change through radiative through radiative forcing and can be derived from both biogenic and anthropogenic sources. Concentrations of these compounds can vary depending on the time of year as well as air mass movement. Both anthropogenic and biogenic OSs were identified in the filter samples collected in Towson, MD. The majority of this OS was biogenic and nature, and derived from isoprene. This is especially true in warm months, and the concentrations show strong

seasonal trends where the total concentration decreases substantially as the seasonal temperature decreases. Biogenic OSs derived from terpene make up approximately 20% of the total OS concentration and show a weaker seasonal trend. Conversely, anthropogenically derived OS show peak concentrations in the winter months and were long chain alkyl species. This is potentially due to the increase of biomass burning for warmth and fuel. Results from the HYSPLIT model show that the movement of air masses also plays a part in the concentration of OS observed on the filters. Air masses that are transported over larger urban centers from Pennsylvania or Virginia tend to have higher OS concentrations, whereas air masses moving in from the Atlantic and the Chesapeake Bay result in low OS concentrations.

VI. Moving Forward

The next step for this research would be to continue to analyze the filters for difference classes of compounds, such as nitrates. By further analyzing the filter samples for these different classes of compounds, it would be possible to gain more understanding of the air mass in general. It would also give insight into the exact percentage each different class of compounds contributes to the total local SOA air mass. Furthermore, after each different class of compounds is analyzed, they could be correlated to each other to see if there is any significant correlation between them. For example, it would be interesting to see the possible correlations between the presence and concentration of different nitrogen species and the presence and concentrations of CHONS that were observed in this study.

The results obtained from the analysis of the collected sample filters also emphasizes the importance and potential implications that a longer sampling duration can have on observed OS levels. Considering the limited sampling duration of other similar OS studies, it could be concluded that perhaps more work should be done to examine OS on a broader timescale. For example, the experiment could be repeated where a new set of samples are collected over the course of a year. These samples could be analyzed as they are collected, rather than doing one bulk analysis at the end of the study. By doing this it would be possible to see if more OS is observed on the sample filters and if prolonged storage has an impact on the concentration of OS present on the filters. By repeating the experiment it would also be possible to examine further correlations with weather conditions as the weather changed on a year to year basis.

Appendix 1-OS Correlations

The following data shows the correlations between each of the observed OS, the trace gas and mass loading values, and the atmospheric conditions. Values that have no color coding have an r-value less than 0.4. Light green values have a positive correlation with an r-value of 0.4 and represent correlation at the 95% level. Dark green values have a positive correlation with an r-value of 0.5 and represent correlation at the 99% level. Light blue values have a negative correlation with an r-value of 0.4 and represent correlation at the 95% level. Dark blue values have a strong negative correlation with an r-value of 0.5, representing correlation at the 95% level. The raw data file can also be examined on order to observe this data more clearly.

		C3H5O5S-	C3H5O6S-	C3H5O6S-	C6H13O4S-
<i>precursor</i>		<i>precursor</i>	<i>isoprene</i>	<i>isoprene</i>	<i>alkyl OS</i>
[M-H]- MW		152.9858	168.9807	168.9807	181.054
C3H5O5S-	<i>precursor</i>	1			
C3H5O6S-	<i>isoprene</i>	0.601719	1		
C3H5O6S-	<i>isoprene</i>	0.688021	0.70349	1	
C6H13O4S-	<i>alkyl OS</i>	-0.2306	-0.25404	-0.23868	1
C4H7O6S-	<i>Isoprene</i>	0.642955	0.16357	0.175494	-0.111062395
C4H7O6S-	<i>alkyl OS;</i>				
C4H7O6S-	<i>Isoprene</i>	0.799541	0.116022	0.32903	-0.125642274
C5H7O6S	<i>Isoprene</i>	0.359662	0.21359	0.362554	-0.056175171
	<i>alkyl OS,</i>				
C5H9O6S-	<i>isoprene</i>	0.621112	0.299037	0.203194	0.034619465
C4H7O7S-	<i>isoprene</i>	0.69502	0.272338	0.290557	-0.123091245
C4H7O7S-	<i>isoprene</i>	0.797593	0.150715	0.347104	-0.122755141
C7H11O5S-	0	0.127259	0.345115	-0.06189	0.416454203
C8H15O4S-	<i>alkyl OS</i>	0.101718	0.379485	0.306709	0.118540094
C7H13O5S-	<i>alkyl OS</i>	0.148577	0.636285	0.266688	0.158710647
C7H13O5S-	<i>alkyl OS</i>	0.081432	0.583572	0.016801	-0.125952972
C6H11O6S-	0	0.269577	0.661675	0.219731	0.102159132
C7H15O5S-	<i>alkyl OS</i>	-0.02692	0.201481	0.0083	-0.170579289

C5H9O7S-	<i>isoprene</i>	0.416407	0.040964	0.060709	0.508160788
C5H9O7S-	<i>isoprene</i>	0.545031	0.035667	0.207776	0.521803749
C5H9O7S-	<i>isoprene</i>	0.495083	-0.10192	0.114986	0.429371721
C5H11O7S-	<i>isoprene</i>	0.233263	0.083404	-0.01914	-0.048101651
C5H11O7S-	<i>isoprene</i>	0.621974	-0.05761	0.152451	0.204499161
C5H11O7S-	<i>sum of 215</i>	0.601088	-0.05228	0.119085	0.301721522
C7H11O6S-		0.053072	0.237979	0.214062	0.179158332
C6H9O7S -	0	0.396225	0.533549	0.349853	0.088295802
C10H16O4S		0.316343	0.480282	0.498785	0.059351515
C9H15O5S-		0.315304	0.647269	0.326099	0.017785028
C7H9O7S-		0.044092	0.184835	0.412257	-0.071425592
C9H17O5S-		-0.09517	0.297729	0.043679	-0.058229168
C7H11O7S-		0.084086	-0.05508	0.048676	0.649612365
C7H11O7S-	<i>limonene</i>	0.271429	0.128895	0.11959	0.648570067
C7H11O7S-	<i>limonene</i>	-0.04763	0.113919	-0.07592	0.521226002
C9H15O6S-	<i>limonene</i>	0.42711	0.507952	0.25966	0.146852717
C6H12NO8S-	0	0.268344	0.453377	0.181424	-0.048222716
C8H11O8S-	<i>isoprene</i>	-0.20368	-0.14642	-0.13478	0.871432737
C9H15O7S-	<i>limonene</i>	0.125665	0.209284	0.316734	0.278576619
C10H15O7S-	<i>α-pinene</i>	0.376304	0.08492	0.154131	0.712028624
C12H23O5S-	0	0.063753	0.468016	0.085545	-0.100286955
C9H15O8S-	<i>α-terpinene</i>	0.001946	-0.1517	0.199307	-0.029154747
C8H14NO8S-	0	-0.11056	-0.22507	0.134044	-0.052474837
C14H29O4S-	0	0.106195	0.098353	0.267681	-0.20252111
C10H16NO7S-	<i>α-pinene</i>	-0.06156	0.352452	-0.0564	-0.076136757
C9H14NO8S-	<i>limonene</i>	0.201429	0.467086	0.212525	-0.004023312
C10H17O8S-	<i>α-pinene</i>	0.455486	-0.10963	0.065449	0.558057437
C13H25O6S-	0	-0.15333	-0.12241	-0.137	-0.035714286
C15H29O5S-	0	0.181498	0.491807	0.338508	0.1879976
C10H16NO9S-	<i>limonene</i>	0.261988	0.382602	0.139766	0.303823417
C10H16NO9S-	<i>terpinolene</i>	0.084274	0.334013	0.010752	0.123211293
C10H16NO9S-	<i>b-pinene</i>	0.257437	0.205377	0.170396	0.622414454
C10H16NO10S-	<i>α-pinene</i>	0.478689	0.372841	0.347625	-0.041348735
C10H16NO10S-	<i>α-terpinene</i>	0.289241	0.425847	0.253884	0.369166667
C10H16NO10S-	<i>b-pinene</i>	0.24707	0.221635	0.18266	0.644746662
sum 342		0.439866	0.382428	0.327376	0.187412862
C16H29O7S	0	0.138531	0.275039	0.389165	-0.074760795
C10H17N2O11S-	<i>limonene</i>	-0.04138	0.116725	-0.13374	0.014105131
C15H16O10S	0	0.143949	0.083478	0.181865	0.436191663
C25H22O7S	0	0.285046	0.348587	0.23544	0.1428853
C22H13NO9S	0	-0.26521	-0.30153	-0.31832	0.885212618
C39H58O4S	0	-0.00382	0.273367	0.443551	-0.038489611
Sulfate Essex		-0.27369	-0.02142	-0.18841	0.399581644
Sulfate Grantsville		0.30381	0.111975	-0.02553	-0.187859179
Sulfate Beltsville		0.24961	0.18007	-0.11491	0.036280329

OC	0.130168	-0.01285	-0.00626	0.281162245
EC	0.224336	0.040646	-0.02647	0.164367003
TOC	0.153135	-0.00089	-0.01092	0.258220957
temp C	0.260112	-0.04992	0.134163	0.274093835
temp high C	0.2841	-0.05843	0.174179	0.258110887
temp low	0.244998	-0.04162	0.04117	0.310016035
RH (%)	0.091827	0.167539	-0.00924	0.115674119
precip (in)	-0.17198	-0.16765	-0.20451	-0.088992159
precip (cm)	-0.15382	-0.17519	-0.20374	-0.081441008
UV-index	0.231838	-0.04872	0.114676	0.301893095
Solar Rad (W*m-2)	0.257762	-0.04044	0.310697	0.096334477
Wind Direction	-0.16579	-0.05689	0.142426	0.14917157
Wind speed	-0.13361	-0.01208	0.08261	-0.136487171

		C4H7O6S-	C4H7O6S-	C5H7O6S
precursor		alkyl OS; Isoprene	alkyl OS; Isoprene	Isoprene
[M-H]- MW		182.99688	182.9969	194.9963
C3H5O5S-	precursor			
C3H5O6S-	isoprene			
C3H5O6S-	isoprene			
C6H13O4S-	alkyl OS			
C4H7O6S-	alkyl OS; Isoprene	1		
C4H7O6S-	alkyl OS; Isoprene	0.84061117	1	
C5H7O6S	Isoprene	0.578278477	0.406945	1
C5H9O6S-	alkyl OS, isoprene	0.762031776	0.571088	0.41072687
C4H7O7S-	isoprene	0.968777407	0.794658	0.652268536
C4H7O7S-	isoprene	0.870622701	0.964907	0.479383408
C7H11O5S-	0	-0.097031341	-0.1622	0.116173648
C8H15O4S-	alkyl OS	-0.051127442	-0.16411	0.084046886
C7H13O5S-	alkyl OS	-0.253178737	-0.27764	0.185171752
C7H13O5S-	alkyl OS	-0.195933308	-0.22993	0.218604902
C6H11O6S-	0	0.006378127	-0.10227	0.102804688
C7H15O5S-	alkyl OS	-0.252432794	-0.19126	0.111819243
C5H9O7S-	isoprene	0.735804904	0.581188	0.59539262
C5H9O7S-	isoprene	0.615670874	0.682409	0.411883958

C5H9O7S-	<i>isoprene</i>	0.414651651	0.699674	0.031295089
C5H11O7S-	<i>isoprene</i>	0.781807198	0.390956	0.689261352
C5H11O7S-	<i>isoprene</i>	0.6259399	0.864736	0.174096197
C5H11O7S-	<i>sum of 215</i>	0.736676352	0.863554	0.311923732
				-
C7H11O6S-		-0.003144612	-0.07002	0.022108099
C6H9O7S -	0	0.054550659	-0.01901	0.026741314
				-
C10H16O4S		-0.219727168	-0.02062	0.073306156
				-
C9H15O5S-		-0.185508806	-0.16519	0.170917168
C7H9O7S-		-0.152176602	-0.11127	0.244066919
				-
C9H17O5S-		-0.116872733	-0.08359	0.135418314
C7H11O7S-		0.207646606	0.123515	0.253899733
C7H11O7S-	<i>limonene</i>	0.154144559	0.136828	0.049777673
				-
C7H11O7S-	<i>limonene</i>	-0.149648392	-0.18577	0.175409691
				-
C9H15O6S-	<i>limonene</i>	0.00491476	0.059012	0.089153141
				-
C6H12NO8S-	0	0.052250888	-0.02853	0.101431657
				-
C8H11O8S-	<i>isoprene</i>	-0.049411787	-0.14004	0.076006975
C9H15O7S-	<i>limonene</i>	-0.087206285	-0.07176	0.065882196
				-
C10H15O7S-	<i>α-pinene</i>	0.158188371	0.336475	0.115990195
				-
C12H23O5S-	0	-0.182683263	-0.20525	0.157809652
				-
C9H15O8S-	<i>α-terpinene</i>	-0.205364622	0.022676	0.057444099
				-
C8H14NO8S-	0	-0.249623351	-0.07908	0.074260826
				-
C14H29O4S-	0	-0.110901462	-0.00752	0.048896491
				-
C10H16NO7S-	<i>α-pinene</i>	-0.174867893	-0.30494	0.204766986
C9H14NO8S-	<i>limonene</i>	0.070967301	-0.09937	0.07908119
				-
C10H17O8S-	<i>α-pinene</i>	0.35810358	0.622599	0.012178136
C13H25O6S-	0	-0.112480081	-0.1276	-0.08665838
C15H29O5S-	0	-0.123647171	-0.17185	0.181271954
				-
C10H16NO9S-	<i>limonene</i>	0.018335796	-0.02787	0.105572419
				-
C10H16NO9S-	<i>terpinolene</i>	0.086867817	-0.04772	0.103292053

C10H16NO9S-	<i>b-pinene</i>	-0.007227089	0.101037	0.124709959	-
C10H16NO10S-	<i>a-pinene</i>	0.140687448	0.214706	0.068587192	-
C10H16NO10S-	<i>a-terpinene</i>	0.038983507	-0.0467	0.004434243	-
C10H16NO10S-	<i>b-pinene</i>	-0.015570759	0.016218	0.076400749	-
sum 342		0.10044108	0.145601	0.066889319	-
C16H29O7S	0	-0.107874122	-0.1001	0.303398495	-
C10H17N2O11S-	<i>limonene</i>	0.197923574	-0.03954	0.040266864	-
C15H16O10S	0	-0.056241684	0.03442	0.003185094	-
C25H22O7S	0	0.067292677	-0.01175	0.026270495	-
C22H13NO9S	0	-0.011801144	-0.10977	0.066459237	-
C39H58O4S	0	-0.163577206	-0.1161	0.043172972	-
Sulfate Essex		-0.193288469	-0.22491	0.089215694	-
Sulfate Grantsville		0.450966447	0.306089	0.121837057	-
Sulfate Beltsville		0.269684221	0.15772	0.029146481	-
OC		0.073765932	0.064695	0.095813914	-
EC		0.236598537	0.143614	0.189510028	-
TOC		0.111585396	0.083387	0.118244393	-
temp C		0.456773893	0.387462	0.496314009	-
temp high C		0.431737967	0.401633	0.49070168	-
temp low		0.45616716	0.354456	0.421980761	-
RH (%)		-0.054267826	-0.05578	0.258157425	-
precip (in)		-0.057937197	-0.11871	0.214115883	-
precip (cm)		-0.045042741	-0.1017	0.199122187	-
UV-index		0.330457582	0.392877	0.436646663	-
Solar Rad (W*m-2)		0.330833512	0.386394	0.407388175	-
Wind Direction		-0.239667889	-0.28279	0.008196467	-
Wind speed		-0.291340668	-0.22478	0.036301162	-

		C5H9O6S-	C4H7O7S-	C4H7O7S-	C7H11O5S-
precursor		alkyl OS, isoprene	isoprene	isoprene	0
[M-H]- MW		197.0125	198.9913	198.9913	207.0333

C3H5O5S-	precursor				
C3H5O6S-	isoprene				
C3H5O6S-	isoprene				
C6H13O4S-	alkyl OS				
	alkyl OS;				
C4H7O6S-	Isoprene				
	alkyl OS;				
C4H7O6S-	Isoprene				
C5H7O6S	Isoprene				
	alkyl OS,				
C5H9O6S-	isoprene	1			
C4H7O7S-	isoprene	0.811612	1		
C4H7O7S-	isoprene	0.645846	0.865003	1	
C7H11O5S-	0	0.300044	-0.03601	-0.13267	1
C8H15O4S-	alkyl OS	0.0366	-0.02312	-0.12981	0.195017
C7H13O5S-	alkyl OS	0.056207	-0.22359	-0.29642	0.586263
C7H13O5S-	alkyl OS	-0.15164	-0.19842	-0.24893	0.425188
C6H11O6S-	0	0.377924	0.073236	-0.07169	0.745237
C7H15O5S-	alkyl OS	-0.3027	-0.29821	-0.25957	0.171741
C5H9O7S-	isoprene	0.680036	0.736564	0.629157	0.284586
C5H9O7S-	isoprene	0.585062	0.626994	0.715964	0.29767
C5H9O7S-	isoprene	0.26709	0.338915	0.658128	0.116125
C5H11O7S-	isoprene	0.603297	0.807292	0.470239	-0.09522
C5H11O7S-	isoprene	0.396843	0.550872	0.838612	-0.01107
C5H11O7S-	sum of 215	0.504123	0.672786	0.855888	0.027424
C7H11O6S-		0.096194	0.0643	-0.00117	0.396637
C6H9O7S -	0	0.604796	0.188972	0.042281	0.666902
C10H16O4S		-0.12878	-0.17718	-0.08716	0.400529
C9H15O5S-		0.183745	-0.07093	-0.1228	0.635279
C7H9O7S-		-0.19275	-0.03246	-0.01629	0.060716
C9H17O5S-		-0.26111	-0.19333	-0.20248	0.027277
C7H11O7S-		0.30594	0.219026	0.089764	0.236872
C7H11O7S-	limonene	0.55616	0.21053	0.121892	0.568423
C7H11O7S-	limonene	0.18706	-0.10386	-0.2005	0.704056
C9H15O6S-	limonene	0.478897	0.104932	0.066128	0.630929
C6H12NO8S-	0	0.560958	0.135737	0.01117	0.462096
C8H11O8S-	isoprene	0.036463	-0.05765	-0.10447	0.341393
C9H15O7S-	limonene	0.119417	-0.05791	-0.0967	0.21289
C10H15O7S-	α -pinene	0.361936	0.163528	0.317173	0.532047
C12H23O5S-	0	-0.16611	-0.1863	-0.21548	0.186024
C9H15O8S-	α -terpinene	-0.32887	-0.21228	-0.08817	-0.17985
C8H14NO8S-	0	-0.34648	-0.27808	-0.16535	-0.21547
C14H29O4S-	0	-0.18563	-0.11354	-0.03385	-0.20665
C10H16NO7S-	α -pinene	-0.01184	-0.18778	-0.30255	0.213371
C9H14NO8S-	limonene	0.271358	0.114974	-0.04761	0.394643

C10H17O8S-	<i>a-pinene</i>	0.320897	0.31766	0.562262	0.221421
C13H25O6S-	0	-0.09638	-0.12374	-0.12393	-0.1339
C15H29O5S-	0	0.169666	-0.01006	-0.11059	0.525105
C10H16NO9S-	<i>limonene</i>	0.485267	0.109678	-0.01184	0.729944
C10H16NO9S-	<i>terpinolene</i>	0.110977	0.036824	-0.04554	0.303574
C10H16NO9S-	<i>b-pinene</i>	0.178588	-0.01818	0.041207	0.416943
C10H16NO10S-	<i>a-pinene</i>	0.470973	0.23268	0.170605	0.318667
C10H16NO10S-	<i>a-terpinene</i>	0.493406	0.1372	0.01022	0.619568
C10H16NO10S-	<i>b-pinene</i>	0.398969	0.064674	0.014384	0.608526
sum 342		0.504575	0.199858	0.123788	0.469927
C16H29O7S	0	-0.14611	0.011906	-0.00644	-0.03051
C10H17N2O11S-	<i>limonene</i>	0.184594	0.133133	-0.02668	0.0861
C15H16O10S	0	0.125461	-0.029	-0.01909	0.263789
C25H22O7S	0	0.575704	0.196601	0.054576	0.589107
C22H13NO9S	0	0.103312	-0.04134	-0.07206	0.326866
C39H58O4S	0	-0.21978	-0.08218	-0.05769	-0.11051
Sulfate Essex		0.052553	-0.2119	-0.22733	0.209345
Sulfate Grantsville		0.523568	0.482687	0.360977	0.005587
Sulfate Beltsville		0.566783	0.331354	0.219341	0.298624
OC		0.308418	0.20945	0.153607	0.261806
EC		0.495369	0.365775	0.252546	0.263556
TOC		0.354642	0.247495	0.177949	0.265483
temp C		0.497367	0.519502	0.455291	0.061371
temp high C		0.498574	0.495976	0.456345	0.002348
temp low		0.532365	0.509567	0.414159	0.177119
RH (%)		0.028342	-0.04977	-0.09876	0.367178
precip (in)		-0.13338	-0.13904	-0.18521	0.114514
precip (cm)		-0.11223	-0.11842	-0.1625	0.128121
UV-index		0.216154	0.402175	0.465127	-0.0699
Solar Rad (W*m-2)		0.20633	0.399812	0.479158	-0.34153
Wind Direction		-0.21554	-0.25247	-0.29638	-0.05781
Wind speed		-0.36834	-0.24899	-0.2331	-0.0446

		C8H15O4S-	C7H13O5S-	C7H13O5S-	C6H11O6S-
<i>precursor</i>		<i>alkyl OS</i>	<i>alkyl OS</i>	<i>alkyl OS</i>	0
[M-H]- MW		207.0697	209.0489	209.0489	211.0276
C8H15O4S-	<i>alkyl OS</i>	1			
C7H13O5S-	<i>alkyl OS</i>	0.626818	1		
C7H13O5S-	<i>alkyl OS</i>	0.427149	0.737422	1	
C6H11O6S-	0	0.371079	0.74412	0.493454	1
C7H15O5S-	<i>alkyl OS</i>	0.17408	0.38579	0.540766	0.017245
C5H9O7S-	<i>isoprene</i>	0.018832	-0.0283	-0.20092	0.114949
C5H9O7S-	<i>isoprene</i>	-0.0306	-0.04552	-0.28853	0.160455
C5H9O7S-	<i>isoprene</i>	-0.09247	-0.08343	-0.18847	-0.02852

C5H11O7S-	isoprene	0.016128	-0.24455	-0.14557	-0.09379
C5H11O7S-	isoprene	-0.13675	-0.19486	-0.20921	-0.09137
C5H11O7S-	sum of 215	-0.10161	-0.20516	-0.23594	-0.08372
C7H11O6S-	0	0.342675	0.284788	0.079808	0.498664
C6H9O7S -		0.281768	0.565943	0.14481	0.804693
C10H16O4S		0.288185	0.596805	0.326503	0.432003
C9H15O5S-		0.435192	0.761987	0.570941	0.762243
C7H9O7S-		0.090832	0.029046	-0.13094	0.157313
C9H17O5S-	limonene	0.300049	0.462766	0.534973	0.309
C7H11O7S-		0.032646	0.025353	-0.20846	0.06816
C7H11O7S-		0.089927	0.278956	-0.14913	0.4703
C7H11O7S-		0.263545	0.52176	0.140922	0.676295
C9H15O6S-		0.159091	0.609739	0.279137	0.749984
C6H12NO8S-	0	0.072611	0.461178	0.116981	0.716481
C8H11O8S-	isoprene	0.159615	0.081836	-0.18901	0.19958
C9H15O7S-	limonene	0.156927	0.254974	0.010987	0.223848
C10H15O7S-	α -pinene	0.117552	0.276251	-0.07795	0.381445
C12H23O5S-	0	0.531691	0.675763	0.870293	0.339209
C9H15O8S-	α -terpinene	-0.09203	-0.16387	-0.3281	-0.1214
C8H14NO8S-	0	-0.1195	-0.17865	-0.37051	-0.18745
C14H29O4S-	0	0.181451	0.169558	0.091358	-0.09938
C10H16NO7S-	α -pinene	0.117972	0.399522	0.412725	0.388042
C9H14NO8S-	limonene	0.191012	0.387166	0.218865	0.613181
C10H17O8S-	α -pinene	-0.0582	-0.02873	-0.19651	0.059233
C13H25O6S-	0	-0.13858	-0.08277	-0.04174	-0.18757
C15H29O5S-	0	0.246275	0.536303	0.157145	0.731212
C10H16NO9S-	limonene	0.167672	0.494834	0.094297	0.830494
C10H16NO9S-	terpinolene	0.288906	0.401577	0.335641	0.550587
C10H16NO9S-	b -pinene	0.444985	0.524453	0.203726	0.413813
C10H16NO10S-	α -pinene	0.016831	0.23329	-0.07512	0.515687
C10H16NO10S-	α -terpinene	0.316244	0.565975	0.101598	0.760709
C10H16NO10S-	b -pinene	0.190898	0.439716	-0.02834	0.565279
sum 342	0	0.109517	0.360992	-0.04249	0.619542
C16H29O7S		0.340866	0.246158	0.149507	0.116203
C10H17N2O11S-		0.038685	0.060421	0.082101	0.322285
C15H16O10S		0.046946	0.142285	-0.05865	0.226676
C25H22O7S		0.114188	0.339282	-0.09777	0.716569
C22H13NO9S	0	0.056207	0.017134	-0.20701	0.106671
C39H58O4S	0	0.047143	-0.06639	-0.06941	-0.05125
Sulfate Essex		-0.0195	0.133878	0.04424	0.165233
Sulfate Grantsville		-0.13851	-0.00993	0.076171	0.119712
Sulfate Beltsville		-0.15462	0.136812	0.194814	0.283451
OC		-0.10309	-0.14454	-0.21941	0.046198
EC		-0.08654	-0.10768	-0.19445	0.106072
TOC		-0.10063	-0.138	-0.2165	0.060343

temp C	-0.28121	-0.25001	-0.56486	0.122035
temp high C	-0.29622	-0.28725	-0.62222	0.08138
temp low	-0.22751	-0.13235	-0.46588	0.245313
RH (%)	-0.02122	0.223997	0.172207	0.305096
precip (in)	-0.21132	0.062542	-0.04606	0.159933
precip (cm)	-0.20746	0.014487	-0.07721	0.142365
UV-index	-0.25503	-0.39387	-0.58175	-0.07558
Solar Rad (W*m-2)	-0.05555	-0.43673	-0.5213	-0.22664
Wind Direction	0.450422	0.193877	0.006385	-0.06767
Wind speed	-0.08518	-0.14047	-0.02392	-0.19602

		C7H15O5S-	C5H9O7S-	C5H9O7S-	C5H9O7S-
precursor		alkyl OS	isoprene	isoprene	isoprene
[M-H]- MW		211.0646	213.0069	213.0069	213.0069
C7H15O5S-	alkyl OS	1			
C5H9O7S-	isoprene	-0.27074	1		
C5H9O7S-	isoprene	-0.32888	0.882447	1	
C5H9O7S-	isoprene	-0.15918	0.571193	0.809025	1
C5H11O7S-	isoprene	-0.20903	0.658185	0.323857	-0.08085
C5H11O7S-	isoprene	-0.18114	0.609015	0.800842	0.951619
C5H11O7S-	sum of 215	-0.22897	0.774438	0.874597	0.909636
C7H11O6S-		-0.17772	0.182542	0.310361	0.04496
C6H9O7S-	0	-0.15797	0.204235	0.261197	-0.0292
C10H16O4S		0.380607	-0.02187	0.127376	0.155661
C9H15O5S-		0.028749	-0.01449	0.090757	-0.03602
C7H9O7S-		-0.13428	-0.00613	0.17473	-0.04936
C9H17O5S-		0.341035	-0.1968	-0.2152	-0.13234
C7H11O7S-		-0.19101	0.573899	0.472624	0.244641
C7H11O7S-	limonene	-0.28863	0.535685	0.575836	0.332251
C7H11O7S-	limonene	-0.17165	0.29219	0.360253	0.134345
C9H15O6S-	limonene	-0.11133	0.184293	0.304653	0.153924
C6H12NO8S-	0	-0.12888	0.047302	0.095518	-0.08092
C8H11O8S-	isoprene	-0.25007	0.41908	0.466815	0.350492
C9H15O7S-	limonene	0.007484	0.135015	0.115034	0.052351
C10H15O7S-	α -pinene	-0.20346	0.56749	0.785848	0.764046
C12H23O5S-	0	0.370471	-0.172	-0.27196	-0.15315
C9H15O8S-	α -terpinene	-0.15735	-0.18835	-0.02152	0.0952
C8H14NO8S-	0	-0.0928	-0.22283	-0.101	0.033959
C14H29O4S-	0	0.09305	-0.13329	-0.04958	-0.0012
C10H16NO7S-	α -pinene	0.053946	-0.17229	-0.23257	-0.20399
C9H14NO8S-	limonene	-0.02334	0.09822	0.061216	-0.08221
C10H17O8S-	α -pinene	-0.22786	0.588308	0.818991	0.906247
C13H25O6S-	0	-0.05658	-0.10303	-0.11942	-0.05991
C15H29O5S-	0	-0.16293	0.13256	0.220919	-0.01173

C10H16NO9S-	<i>limonene</i>	-0.23477	0.249179	0.355642	0.113277
C10H16NO9S-	<i>terpinolene</i>	-0.00115	0.104865	0.102517	0.099231
C10H16NO9S-	<i>b-pinene</i>	-0.15391	0.363492	0.449559	0.460639
C10H16NO10S-	<i>a-pinene</i>	-0.2857	0.091063	0.239248	0.066741
C10H16NO10S-	<i>a-terpinene</i>	-0.2938	0.325913	0.385612	0.147524
C10H16NO10S-	<i>b-pinene</i>	-0.28113	0.420633	0.532304	0.332607
sum 342		-0.31464	0.218252	0.35805	0.150915
C16H29O7S	0	-0.00404	0.030269	0.089344	-0.06933
C10H17N2O11S-	<i>limonene</i>	-0.09877	0.060624	-0.05173	-0.10456
C15H16O10S	0	-0.08272	0.206896	0.264627	0.242781
C25H22O7S	0	-0.30233	0.218202	0.324547	0.007501
C22H13NO9S	0	-0.24918	0.468126	0.454131	0.346047
C39H58O4S	0	-0.0192	-0.16624	-0.05768	-0.06382
Sulfate Essex		-0.25626	0.219592	0.097045	0.04038
Sulfate Grantsville		-0.25382	0.109765	0.07696	0.010555
Sulfate Beltsville		-0.2718	0.21343	0.144372	0.061328
OC		-0.38111	0.312864	0.364407	0.182006
EC		-0.44347	0.393572	0.376126	0.128815
TOC		-0.40002	0.335071	0.371628	0.172234
temp C		-0.57044	0.600119	0.614051	0.267686
temp high C		-0.59049	0.555493	0.593875	0.283818
temp low		-0.5373	0.619612	0.625971	0.26044
RH (%)		-0.03239	0.080531	0.086456	0.116039
precip (in)		-0.01601	-0.05005	-0.04452	-0.09913
precip (cm)		-0.05577	-0.03259	-0.02138	-0.08656
UV-index		-0.39105	0.413725	0.511379	0.351991
Solar Rad (W*m-2)		-0.3571	0.286726	0.371202	0.303516
Wind Direction		0.26228	-0.11823	-0.16802	-0.13224
Wind speed		0.3395	-0.32509	-0.2838	-0.17629

		C5H11O7S-	C5H11O7S-	C5H11O7S-	C7H11O6S-
<i>precursor</i>		<i>isoprene</i>	<i>isoprene</i>	<i>sum of 215</i>	
[M-H]- MW		215.0226	215.0226	215.0226	223.0282
C5H11O7S-	<i>isoprene</i>	1			
C5H11O7S-	<i>isoprene</i>	0.118929	1		
C5H11O7S-	<i>sum of 215</i>	0.328846	0.968082	1	
C7H11O6S-		0.020622	-0.00591	0.028493	1
C6H9O7S -	0	-0.01017	-0.07183	-0.04895	0.35463
C10H16O4S		-0.33975	0.052488	-0.00405	0.327376
C9H15O5S-		-0.22517	-0.1047	-0.13588	0.469004
C7H9O7S-		-0.09621	-0.0715	-0.08754	0.710178
C9H17O5S-		-0.12404	-0.15068	-0.17427	0.154554
C7H11O7S-		0.291609	0.133892	0.282238	0.191917
C7H11O7S-	<i>limonene</i>	0.069956	0.195068	0.283339	0.227446

C7H11O7S-	limonene	-0.12048	-0.02337	0.024024	0.586461
C9H15O6S-	limonene	-0.15221	0.086339	0.06978	0.228224
C6H12NO8S-	0	-0.05858	-0.08833	-0.09939	0.099686
C8H11O8S-	isoprene	-0.04359	0.147789	0.237003	0.321999
C9H15O7S-	limonene	-0.0876	-0.0637	-0.02641	0.245981
C10H15O7S-	α -pinene	-0.15995	0.603698	0.612089	0.348619
C12H23O5S-	0	-0.12956	-0.17592	-0.19782	0.053824
C9H15O8S-	α -terpinene	-0.28989	0.030431	-0.02964	0.118436
C8H14NO8S-	0	-0.29562	-0.0294	-0.08943	0.083713
C14H29O4S-	0	-0.16791	0.002554	-0.05402	0.068098
C10H16NO7S-	α -pinene	-0.14763	-0.24858	-0.26126	0.068249
C9H14NO8S-	limonene	-0.02086	-0.11473	-0.09883	0.140852
C10H17O8S-	α -pinene	-0.05484	0.832263	0.817816	0.056837
C13H25O6S-	0	-0.04809	-0.06601	-0.0754	-0.07551
C15H29O5S-	0	-0.16133	-0.10707	-0.10295	0.314866
C10H16NO9S-	limonene	-0.09044	0.014188	0.037901	0.383216
C10H16NO9S-	terpinolene	-0.09368	0.042677	0.041114	0.180952
C10H16NO9S-	β -pinene	-0.18361	0.287433	0.309089	0.187596
C10H16NO10S-	α -pinene	-0.04979	0.061432	0.045403	0.111163
C10H16NO10S-	α -terpinene	-0.05384	0.040792	0.079314	0.326048
C10H16NO10S-	β -pinene	-0.11335	0.160665	0.209962	0.291631
sum 342		-0.07082	0.088222	0.095286	0.201068
C16H29O7S	0	-0.0219	-0.07152	-0.07434	0.292212
C10H17N2O11S-	limonene	0.110307	-0.09794	-0.06142	0.002282
C15H16O10S	0	-0.14059	0.096389	0.125909	0.15851
C25H22O7S	0	-0.01068	-0.0382	-0.01526	0.355292
C22H13NO9S	0	0.014801	0.163074	0.259849	0.132157
C39H58O4S	0	-0.11458	-0.09054	-0.10919	0.310124
Sulfate Essex		-0.07499	-0.05822	-0.00729	-0.03795
Sulfate Grantsville		0.73	0.133794	0.114609	-0.11359
Sulfate Beltsville		0.41737	0.10495	0.109779	-0.1164
OC		0.150289	0.139748	0.205381	0.136007
EC		0.31036	0.147493	0.231593	0.072041
TOC		0.188442	0.143254	0.213893	0.123218
temp C		0.389966	0.307792	0.399573	0.168895
temp high C		0.332004	0.316769	0.393267	0.083126
temp low		0.399428	0.292288	0.39225	0.259063
RH (%)		-0.09902	0.024781	0.041049	0.157192
precip (in)		-0.07845	-0.11258	-0.12648	0.364947
precip (cm)		-0.06851	-0.09857	-0.11024	0.3701
UV-index		0.240741	0.379804	0.427299	-0.00922
Solar Rad (W*m-2)		0.206186	0.344539	0.374653	-0.08719
Wind Direction		-0.14894	-0.22854	-0.21442	-0.05465
Wind speed		-0.18386	-0.20475	-0.23982	-0.17491

		C6H9O7S			
		-	C10H16O4S	C9H15O5S-	C7H9O7S-
precursor		0			
[M-H]- MW		225.008	231.071	235.06405	237.0069
C6H9O7S -	0	1			
C10H16O4S		0.350831	1		
C9H15O5S-		0.737983	0.520277	1	
C7H9O7S-		0.141868	0.39002	0.314282741	1
C9H17O5S-		-0.08594	0.204901	0.094754184	-0.11313
				-	
C7H11O7S-		0.157374	0.083535	0.077650358	-0.08053
C7H11O7S-	limonene	0.661076	0.190188	0.321386361	-0.11456
C7H11O7S-	limonene	0.632034	0.477991	0.602742106	0.25345
C9H15O6S-	limonene	0.87905	0.356328	0.818781556	0.030089
C6H12NO8S-	0	0.852303	0.11999	0.548180063	-0.09644
				-	
C8H11O8S-	isoprene	0.080001	-0.00511	0.005136291	0.094437
C9H15O7S-	limonene	0.242439	0.352787	0.140110657	0.093511
C10H15O7S-	α -pinene	0.429585	0.364901	0.358147601	0.058076
C12H23O5S-	0	0.070946	0.259559	0.512544609	-0.09015
				-	
C9H15O8S-	α -terpinene	-0.11856	0.358633	0.108213656	0.293668
				-	
C8H14NO8S-	0	-0.1709	0.339237	0.194958449	0.292159
C14H29O4S-	0	0.008015	0.246737	0.288717873	0.269032
C10H16NO7S-	α -pinene	0.206739	0.019147	0.472819395	-0.04162
C9H14NO8S-	limonene	0.486263	0.188781	0.429546676	0.135723
C10H17O8S-	α -pinene	0.117937	0.166355	0.067291968	-0.10653
C13H25O6S-	0	-0.12984	-0.33804	0.095781336	-0.07143
C15H29O5S-	0	0.682233	0.447555	0.631513456	0.431359
C10H16NO9S-	limonene	0.894712	0.346833	0.678814452	0.102524
C10H16NO9S-	terpinolene	0.233338	0.054263	0.356256671	0.022954
C10H16NO9S-	b-pinene	0.390692	0.390394	0.465524696	-0.09579
C10H16NO10S-	α -pinene	0.743744	0.260874	0.504008796	0.00299
C10H16NO10S-	α -terpinene	0.851939	0.302206	0.708914543	0.141248
C10H16NO10S-	b-pinene	0.713748	0.346509	0.547374558	0.057665
sum 342		0.82893	0.31455	0.598630901	0.039096
C16H29O7S	0	0.14256	0.352133	0.416299114	0.686577
				-	
C10H17N2O11S-	limonene	0.064183	-0.21466	0.062190259	-0.16163
C15H16O10S	0	0.248595	0.335659	0.130202745	-0.0056
C25H22O7S	0	0.92825	0.215942	0.599030329	0.204171
				-	
C22H13NO9S	0	0.040976	-0.12973	0.121760823	-0.11958

C39H58O4S	0	-0.09741	0.22069	0.009627762	0.417208
Sulfate Essex		0.111061	-0.30276	0.010731541	-0.16069
Sulfate Grantsville		0.154121	-0.43784	0.096474256	-0.25774
Sulfate Beltsville		0.335726	-0.45458	0.239046563	-0.33513
OC		0.352882	-0.14132	0.263311684	0.1296
EC		0.446216	-0.23848	0.308141757	0.042424
TOC		0.378451	-0.16511	0.276768447	0.111471
temp C		0.264834	-0.05358	0.090407994	0.243222
temp high C		0.276648	-0.09901	0.124868031	0.199439
temp low		0.337289	-0.01423	0.029901768	0.222896
RH (%)		0.290894	0.351476	0.331534526	0.030222
precip (in)		0.002882	0.210214	0.07786953	0.30008
precip (cm)		0.014584	0.186021	0.083555962	0.308925
UV-index		-0.06427	-0.13515	0.262214055	0.13314
Solar Rad (W*m-2)		-0.06703	-0.23665	0.337019186	0.122516
Wind Direction		-0.04132	0.053433	0.139866662	0.040731
Wind speed		-0.19525	0.231352	0.187648301	0.106471

		C9H17O5S-	C7H11O7S-	C7H11O7S-	C7H11O7S-
precursor				limonene	limonene
[M-H]- MW		237.0802	239.0226	239.022552	239.0226
C9H17O5S-		1			
C7H11O7S-		-0.13201	1		
C7H11O7S-	limonene	-0.16009	0.760498	1	
C7H11O7S-	limonene	0.14127	0.378077	0.65672294	1
C9H15O6S-	limonene	0.020145	0.107641	0.634654613	0.634368
C6H12NO8S-	0	0.070539	-0.12454	0.459125466	0.422329
C8H11O8S-	isoprene	-0.10952	0.589154	0.575827644	0.431268
C9H15O7S-	limonene	-0.14348	0.722836	0.51206836	0.26737
C10H15O7S-	α -pinene	-0.12302	0.534727	0.777819607	0.621769
C12H23O5S-	0	0.395057	-0.06654	-0.1318546	0.094827
C9H15O8S-	α -terpinene	-0.01514	0.1811	0.036250872	0.152619
C8H14NO8S-	0	-0.08645	0.092507	-0.07210802	0.036731
C14H29O4S-	0	0.061803	-0.26942	-0.1891259	-0.03584
C10H16NO7S-	α -pinene	0.03448	-0.08916	0.029203672	0.141671
C9H14NO8S-	limonene	-0.12807	0.102924	0.298343169	0.283469
C10H17O8S-	α -pinene	-0.10141	0.366118	0.546619987	0.340725
C13H25O6S-	0	-0.1133	-0.10141	-0.12263484	-0.14986
C15H29O5S-	0	0.015779	0.029741	0.390187059	0.571792

C10H16NO9S-	<i>limonene</i>	0.016071	0.230471	0.74381594	0.799367
C10H16NO9S-	<i>terpinolene</i>	0.147571	0.036617	0.18560505	0.266016
C10H16NO9S-	<i>b-pinene</i>	0.056309	0.578275	0.708658863	0.592072
C10H16NO10S-	<i>a-pinene</i>	-0.02125	0.107073	0.596393358	0.469108
C10H16NO10S-	<i>a-terpinene</i>	-0.15447	0.295606	0.723326332	0.690605
C10H16NO10S-	<i>b-pinene</i>	-0.14355	0.565428	0.911567021	0.758348
sum 342		-0.07472	0.259436	0.753267253	0.622368
C16H29O7S	0	-0.15258	-0.09527	-0.12208525	0.148971
C10H17N2O11S-	<i>limonene</i>	-0.01723	0.180388	0.151506643	0.018664
C15H16O10S	0	-0.13422	0.778213	0.660338989	0.403928
C25H22O7S	0	-0.15376	0.099968	0.65069994	0.638694
C22H13NO9S	0	-0.13943	0.616068	0.59230791	0.409447
C39H58O4S	0	0.051709	-0.14502	-0.20844949	-0.11939
Sulfate Essex		0.196438	0.1142	0.212992475	0.178812
Sulfate Grantsville		-0.13849	-0.09806	0.077320606	-0.19459
Sulfate Beltsville		-0.13212	0.011952	0.284466905	-0.02398
OC		-0.37302	0.265187	0.427962214	0.337949
EC		-0.42214	0.202451	0.41893439	0.292705
TOC		-0.38882	0.254295	0.431278524	0.331932
temp C		-0.40326	0.429541	0.463594554	0.288146
temp high C		-0.43965	0.420679	0.484659304	0.223635
temp low		-0.33418	0.46017	0.530084189	0.423607
RH (%)		-0.06827	0.250873	0.314171189	0.411799
precip (in)		0.034084	0.014437	-0.01912809	0.313838
precip (cm)		-0.05555	0.033697	0.003637452	0.313764
UV-index		-0.23109	0.27551	0.224594217	-0.00219
Solar Rad (W*m-2)		-0.32086	0.215876	0.104403219	-0.21708
Wind Direction		0.016	0.180101	0.03472129	-0.07895
Wind speed		-0.01185	-0.22763	-0.28394605	-0.21903

		C9H15O6S-	C6H12NO8S-	C8H11O8S-	C9H15O7S-
<i>precursor</i>		<i>limonene</i>	0	<i>isoprene</i>	<i>limonene</i>
[M-H]- MW		251.0589	258.0289	267.01743	267.0539
C9H15O6S-	<i>limonene</i>	1			
C6H12NO8S-	0	0.802444	1		
C8H11O8S-	<i>isoprene</i>	0.055294	-0.04183	1	
C9H15O7S-	<i>limonene</i>	0.129186	-0.06892	0.323447163	1
C10H15O7S-	<i>a-pinene</i>	0.536696	0.248417	0.641480092	0.347498
				-	
C12H23O5S-	0	0.206576	-0.03767	0.160205006	0.233267
C9H15O8S-	<i>a-terpinene</i>	-0.0883	-0.29164	0.010473019	0.306018
				-	
C8H14NO8S-	0	-0.21086	-0.30045	0.002788159	0.285049

C14H29O4S-	0	0.14383	-0.00856	0.231814365	-0.21135
C10H16NO7S-	<i>a-pinene</i>	0.326736	0.272117	0.057863341	0.125724
C9H14NO8S-	<i>limonene</i>	0.413771	0.399392	0.285291763	0.343253
C10H17O8S-	<i>a-pinene</i>	0.323119	0.046261	0.440273231	0.054885
C13H25O6S-	0	0.120067	-0.04822	0.057146396	-0.12712
C15H29O5S-	0	0.634336	0.534447	0.289135798	0.146879
C10H16NO9S-	<i>limonene</i>	0.872419	0.791251	0.314578503	0.153607
C10H16NO9S-	<i>terpinolene</i>	0.285782	0.245176	0.415949598	0.14744
C10H16NO9S-	<i>b-pinene</i>	0.507483	0.13668	0.535588622	0.498493
C10H16NO10S-	<i>a-pinene</i>	0.771607	0.720964	-0.01811834	0.012792
C10H16NO10S-	<i>a-terpinene</i>	0.834384	0.686633	0.420417776	0.327202
C10H16NO10S-	<i>b-pinene</i>	0.758591	0.517727	0.595894412	0.404887
sum 342		0.856279	0.738697	0.200131713	0.160226
C16H29O7S	0	0.135672	-0.10094	0.001166719	0.097512
C10H17N2O11S-	<i>limonene</i>	-0.03793	0.110478	0.366157774	0.279659
C15H16O10S	0	0.244598	-0.04276	0.429011683	0.86491
C25H22O7S	0	0.814226	0.860179	0.190000828	0.059679
C22H13NO9S	0	0.027193	-0.04857	0.911844421	0.276978
C39H58O4S	0	-0.19511	-0.07376	0.091913577	0.05131
Sulfate Essex		0.137173	0.227101	0.27697782	-0.04846
Sulfate Grantsville		0.178847	0.230782	0.141298289	0.009187
Sulfate Beltsville		0.387802	0.404942	-0.00461864	0.000371
OC		0.398339	0.255537	0.14540477	-0.02766
EC		0.465587	0.368901	0.038535079	-0.10637
TOC		0.418567	0.284425	0.123011569	-0.04584
temp C		0.162508	0.156702	0.333744082	0.188602
temp high C		0.173383	0.197781	0.31568327	0.195303
temp low		0.275824	0.212174	0.363755755	0.188317
RH (%)		0.322023	0.169623	0.105856737	0.205788
precip (in)		-0.03958	-0.04865	0.088618849	0.079329
precip (cm)		-0.03293	-0.06421	0.098520365	0.092994
UV-index		-0.06084	-0.12877	0.339188365	0.033864
Solar Rad (W*m-2)		-0.13926	-0.11125	0.171075331	0.077669
Wind Direction		-0.19889	-0.1836	0.229887407	0.284434
Wind speed		-0.26832	-0.18429	-0.13229609	-0.15736

		C12H23O5S	C8H14NO8S
		C10H15O7S-	C9H15O8S-
		-	-
precursor		<i>a-pinene</i>	<i>a-terpinene</i>
[M-H] ⁻ MW		279.053852	283.048767
C10H15O7S-	<i>a-pinene</i>	1	284.0446

C12H23O5S-	0	0.071531293	1		
C9H15O8S-	<i>α-terpinene</i>	0.065861357	-0.21965	1	
C8H14NO8S-	0	0.024084824	-0.25724	0.890629136	1
C14H29O4S-	0	0.033572949	0.189522	-0.035928021	0.103558
C10H16NO7S-	<i>α-pinene</i>	0.012673123	0.345334	-0.119798847	-0.14692
C9H14NO8S-	<i>limonene</i>	0.174469264	0.242487	-0.021503311	-0.03588
C10H17O8S-	<i>α-pinene</i>	0.852825805	-0.16614	0.103875276	-0.04137
C13H25O6S-	0	0.084731374	0.015306	8.47213E-05	-0.02307
C15H29O5S-	0	0.319979456	0.119764	0.12671004	0.084399
C10H16NO9S-	<i>limonene</i>	0.588276299	-0.04779	0.007645471	-0.10415
C10H16NO9S-	<i>terpinolene</i>	0.244564935	0.331132	-0.128259755	-0.16792
C10H16NO9S-	<i>b-pinene</i>	0.758920905	0.329445	0.129040433	3.92E-06
C10H16NO10S-	<i>α-pinene</i>	0.385685018	-0.16615	0.133337248	-0.04065
C10H16NO10S-	<i>α-terpinene</i>	0.604877557	0.149549	-0.051374094	-0.11647
C10H16NO10S-	<i>b-pinene</i>	0.81786121	0.006179	0.06502892	-0.05168
sum 342		0.564734552	-0.08961	0.10005082	-0.05979
C16H29O7S	0	0.000657816	0.376012	0.10231925	0.083037
C10H17N2O11S					
-	<i>limonene</i>	0.004171623	0.133691	-0.153183531	-0.14975
C15H16O10S	0	0.537140917	0.135383	0.35236065	0.252213
C25H22O7S	0	0.440897748	-0.21381	-0.075343518	-0.13344
C22H13NO9S	0	0.599989975	-0.16789	-0.075924926	-0.08656
C39H58O4S	0	0.017044049	-0.10682	0.04430488	0.082839
Sulfate Essex		0.124785656	-0.00596	-0.076544644	-0.05159
Sulfate Grantsville		0.030324661	0.136361	-0.392926761	-0.37317
Sulfate Beltsville		0.128319709	0.14411	-0.418797572	-0.43364
OC		0.356334904	-0.17583	-0.130178851	-0.3039
EC		0.278947725	-0.15578	-0.218880749	-0.34
TOC		0.343264673	-0.17349	-0.151908271	-0.31589
temp C		0.355883139	-0.53832	0.128852152	0.038512
temp high C		0.353912061	-0.58415	0.116123821	0.01924
temp low		0.409991983	-0.45653	0.163526659	0.06631
RH (%)		0.304964745	0.081305	0.226466052	0.196385
precip (in)		0.004653186	-0.09844	0.262288487	0.331171
precip (cm)		0.021083091	-0.11575	0.276503401	0.344689
UV-index		0.270755643	-0.467	0.092301384	-0.08993
Solar Rad (W*m-2)		0.132058004	-0.42293	0.067114856	-0.10542

Wind Direction	0.077638385	0.171251	0.132647667	0.121435
Wind speed	-0.2641784	-0.18745	0.192988655	0.19729

		<i>C14H29O4S</i>	<i>C10H16NO7S-</i>	<i>C9H14NO8S</i>	<i>C10H17O8S-</i>
<i>precursor</i>		0	<i>α-pinene</i>	<i>limonene</i>	<i>α-pinene</i>
[M-H] ⁻ MW		293.1788	294.064751	296.044	297.064417
<i>C14H29O4S-</i>	0	1			
<i>C10H16NO7S-</i>	<i>α-pinene</i>	0.080947	1		
<i>C9H14NO8S-</i>	<i>limonene</i>	-0.03454	0.503959756	1	
<i>C10H17O8S-</i>	<i>α-pinene</i>	-0.04605	-0.242430363	-0.06273	1
<i>C13H25O6S-</i>	0	0.227888	0.373722425	0.020421	-0.08012352
<i>C15H29O5S-</i>	0	0.042283	0.248555838	0.656961	0.07187417
					0.31663638
<i>C10H16NO9S-</i>	<i>limonene</i>	-0.07117	0.244718649	0.489161	3
<i>C10H16NO9S-</i>	<i>terpinolene</i>	0.033994	0.615262727	0.839434	0.0825813
					0.57359077
<i>C10H16NO9S-</i>	<i>b-pinene</i>	0.057955	0.250744356	0.285821	2
					0.31598459
<i>C10H16NO10S-</i>	<i>α-pinene</i>	0.139133	0.102960947	0.33079	2
					0.30646331
<i>C10H16NO10S-</i>	<i>α-terpinene</i>	0.00744	0.30188232	0.66126	8
					0.57008381
<i>C10H16NO10S-</i>	<i>b-pinene</i>	-0.02136	0.115649492	0.417695	5
sum 342		0.092808	0.14887657	0.439039	0.40761513
					-
					0.05228842
<i>C16H29O7S</i>	0	0.368722	0.007768377	0.304172	7
					-
<i>C10H17N2O11S</i>					0.11420854
-	<i>limonene</i>	-0.25303	0.366350102	0.743949	7
					0.35193614
<i>C15H16O10S</i>	0	-0.24096	0.007064286	0.273852	8
<i>C25H22O7S</i>	0	-0.00383	0.155194268	0.455956	0.19071328
<i>C22H13NO9S</i>	0	-0.29881	-0.000563047	0.220987	0.43906982
					-
					0.10618854
<i>C39H58O4S</i>	0	0.114373	-0.157755263	-0.11864	1
					0.06656746
Sulfate Essex		-0.27297	0.291383731	-0.08726	8
					0.05961687
Sulfate Grantsville		-0.10839	0.140034191	0.177006	8
					0.10998232
Sulfate Beltsville		-0.25306	0.386844209	0.162845	8
OC		-0.05949	-0.002588457	-0.11225	0.3051347

EC	-0.01607	0.108277734	-0.00874	0.23042303	2
TOC	-0.0504	0.022499611	-0.0902	0.29202921	8
temp C	-0.34132	-0.26660547	0.27718	0.36982048	3
temp high C	-0.345	-0.278551638	0.251857	0.38763005	7
temp low	-0.32657	-0.159527673	0.324377	0.39052309	4
RH (%)	0.112994	0.324376673	0.268471	0.08392722	3
precip (in)	0.232785	0.231592312	0.368691	0.11553139	7
precip (cm)	0.245321	0.204914306	0.373108	0.09828851	4
UV-index	-0.53244	-0.44949877	-0.01185	0.45873038	0.24965740
Solar Rad (W*m-2)	-0.33025	-0.346209139	-0.02519	-0.16088641	6
Wind Direction	-0.07134	-0.161299355	0.140897	-	-
Wind speed	-0.12591	-0.330023344	-0.19594	0.18672052	6

		C13H25O6S	C15H29O5S-	C10H16NO9S	C10H16NO9S
		-	-	-	-
precursor		0	0	limonene	terpinolene
[M-H]- MW		309.1373	321.17367	326.0546	326.054581
C13H25O6S-	0	1			
C15H29O5S-	0	-0.13709	1		
C10H16NO9S-	limonene	-0.08613	0.72227732	4	1
C10H16NO9S-	terpinolene	0.10699	0.48713659	0.366939	7
C10H16NO9S-	b-pinene	-0.09254	0.39527585	0.503828	4
C10H16NO10S-	a-pinene	-0.07311	0.46034654	0.786726	1
C10H16NO10S-	a-terpinene	0.032354	0.79655563	0.866932	8
C10H16NO10S-	b-pinene	-0.0521	0.59251281	0.818999	8
sum 342		-0.05815	0.59349720	0.886712	5

C16H29O7S	0	-0.07476	0.50393765 6	0.020262	0.176200857
C10H17N2O11S			0.21145660		
-	limonene	-0.11454	1	0.138561	0.780054683
			0.19009521		
C15H16O10S	0	-0.12197	7	0.31782	0.137954378
			0.68961962		
C25H22O7S	0	-0.08194	9	0.927013	0.240388246
			0.18463769		
C22H13NO9S	0	-0.05979	7	0.267643	0.342344107
C39H58O4S	0	-0.07342	-0.05639594	-0.15146	-0.200889034
Sulfate Essex		#DIV/0!	0.19221402	0.174243	0.015864024
Sulfate Grantsville		#DIV/0!	-0.04242624	0.017522	0.13273118
			0.08652788		
Sulfate Beltsville		#DIV/0!	3	0.230804	0.129246869
			0.09219397		
OC		0.083493	3	0.367849	-0.195173322
			0.13948829		
EC		0.107285	6	0.405944	-0.11797089
			0.10406518		
TOC		0.08993	4	0.381089	-0.180125786
			0.39353672		
temp C		-0.20781	1	0.316566	0.093913736
			0.38373672		
temp high C		-0.23596	2	0.307712	0.073887656
temp low		-0.13076	0.43608117	0.431609	0.166752146
			0.17579703		
RH (%)		-0.05529	5	0.387509	0.229725037
			0.11458246		
precip (in)		-0.08899	4	0.102187	0.410090544
			0.11860720		
precip (cm)		-0.08144	3	0.110609	0.394754549
			0.20047089		
UV-index		-0.2204	8	0.049089	-0.085571153
			0.04767016		
Solar Rad (W*m-2)		-0.04204	8	-0.08629	-0.081167696
Wind Direction		-0.15709	0.10480181	-0.1534	0.120613826
Wind speed		-0.13649	-0.04563484	-0.17418	-0.328650535

		C10H16NO9S-	C10H16NO10S-	C10H16NO10S-	C10H16NO10S-
precursor		b-pinene	a-pinene	a-terpinene	b-pinene
[M-H]- MW		326.054581	342.049496	342.049496	342.049496
C10H16NO9S-	b-pinene	1			
C10H16NO10S-	a-pinene	0.327613262	1		
C10H16NO10S-	a-terpinene	0.620390938	0.625179905	1	
C10H16NO10S-	b-pinene	0.755446433	0.656364613	0.867795569	1

sum 342		0.511797595	0.956216281	0.807906577	0.839951728
C16H29O7S	0	0.132021601	-0.003943962	0.290135416	0.143038856
C10H17N2O11S-	limonene	0.134944997	0.031633716	0.279648613	0.117416989
C15H16O10S	0	0.623740959	0.190810146	0.393430562	0.569498279
C25H22O7S	0	0.277570896	0.799268867	0.81674135	0.712781717
C22H13NO9S	0	0.525329135	-0.067385916	0.355529745	0.549805344
C39H58O4S	0	-0.271223822	-0.104415053	-0.137060397	-0.108108102
Sulfate Essex		0.149112295	-0.053829635	0.157947836	0.18460217
Sulfate Grantsville		-0.016490995	0.125244737	0.136382967	0.020013862
Sulfate Beltsville		0.127840378	0.204962481	0.277676399	0.215023061
OC		0.191986598	0.353977277	0.283563064	0.421181247
EC		0.163236936	0.410738773	0.353780855	0.39746362
TOC		0.187877752	0.371273444	0.303026002	0.421084125
temp C		0.170748986	0.280125325	0.428228254	0.436023189
temp high C		0.189564769	0.321549899	0.424676981	0.445621168
temp low		0.242756583	0.338113525	0.512056969	0.509995885
RH (%)		0.33986797	0.318044843	0.283964038	0.344983636
precip (in)		-0.022149316	0.100142572	0.069847078	0.057826068
precip (cm)		-0.004438021	0.111126902	0.084787841	0.072698232
UV-index		0.081534368	0.027925395	0.102342088	0.200466759
Solar Rad (W*m-2)		0.032632811	-0.023092135	0.025666358	0.037853671
Wind Direction		0.139199999	-0.195461853	0.046964424	0.044062992
Wind speed		-0.354216043	-0.112780061	-0.323198931	-0.252966547

		C16H29O7S	C10H17N2O11S-	C15H16O10S	C25H22O7S
precursor		0	limonene	0	0
[M-H]- MW		365.16347	373.055309	387.0362	465.1022
C16H29O7S	0	1			
C10H17N2O11S-	limonene	-0.052753697	1		
C15H16O10S	0	0.050552833	0.20772029	1	
C25H22O7S	0	0.085786557	0.077887083	0.160106543	1
C22H13NO9S	0	-0.122052559	0.365563127	0.406295895	0.136416218
C39H58O4S	0	0.096325364	-0.205090198	-0.064573744	-0.054921502
Sulfate Essex		-0.191859132	-0.082536294	-0.037055657	0.142878451
Sulfate Grantsville		-0.098940674	0.323424262	-0.101434678	0.093668626
Sulfate Beltsville		-0.21708076	0.213507741	-0.084945091	0.254698709
OC		0.119850204	-0.34861808	0.174795423	0.421044305
EC		0.12035731	-0.25059673	0.037077206	0.491102147
TOC		0.121466688	-0.330775797	0.145780667	0.442193481
temp C		0.338251899	0.253300038	0.265808306	0.368008021
temp high C		0.320966333	0.235531755	0.279655522	0.3848571
temp low		0.293551793	0.275601626	0.290433717	0.434267792
RH (%)		-0.105011674	0.084390684	0.264104603	0.234937943
precip (in)		0.149521497	0.425211933	-0.061285748	0.048696361

precip (cm)	0.160640789	0.419416814	-0.037558644	0.06385126
UV-index	0.271781409	0.042856129	0.245297054	0.053602426
Solar Rad (W*m-2)	0.240452885	0.005238057	0.182101195	0.006202822
Wind Direction	0.256828765	0.180717019	0.137438736	-0.131576087
Wind speed	-0.0055622	-0.3201889	-0.089464138	-0.139567404

		C22H13NO9S	C39H58O4S	Sulfate Essex	Sulfate Grantsville
<i>precursor</i>		0	0		
[M-H] ⁻ MW		466.027	621.394		
C22H13NO9S	0	1			
		-			
C39H58O4S	0	0.116392636	1		
Sulfate Essex		0.28765589	0.020381094	1	
		-			
Sulfate Grantsville		0.087278071	-0.22245948	-0.02426	1
Sulfate Beltsville		0.07045013	-0.27593136	0.417901	0.812073
OC		0.181968525	0.009635426	0.255799	-0.23737
EC		0.122555755	-0.17876014	0.301909	-0.05491
TOC		0.170786402	-0.03293126	0.268762	-0.19951
temp C		0.411265276	-0.14033006	0.043888	0.216017
temp high C		0.379250553	-0.15663796	0.075042	0.234626
temp low		0.435944987	-0.25942427	0.039519	0.229086
RH (%)		0.111811238	-0.1072631	0.048179	-0.47231
precip (in)		0.061292152	-0.15729075	-0.32158	0.001405
precip (cm)		0.071984642	-0.15427141	-0.3001	0.029568
UV-index		0.361108326	0.054267176	0.040903	0.19619
Solar Rad (W*m-2)		0.217845538	0.067609248	-0.10196	-0.14401
Wind Direction		0.185745375	-0.06718645	0.023061	-0.30969
		-			
Wind speed		0.228498284	0.299371262	-0.2731	-0.52388

		Sulfate Beltsville	OC	EC	TOC	temp C	temp high C
<i>precursor</i>							
[M-H] ⁻ MW							
Sulfate Beltsville		1					
OC		0.123193	1				
EC		0.344955	0.929205	1			
TOC		0.173631	0.996488	0.956888	1		
temp C		0.156577	0.220973	0.311709	0.24424	1	
temp high C		0.188179	0.285345	0.355551	0.304801	0.973253	1

temp low	0.195275	0.195844	0.320884	0.226526	0.960684	0.898568
RH (%)	-0.14392	0.325106	0.370278	0.339423	-0.01819	-0.06184
precip (in)	-0.12947	-0.47435	-0.34944	-0.45272	0.170871	0.086214
precip (cm)	-0.09668	-0.43681	-0.31308	-0.41455	0.179653	0.098756
UV-index	0.118646	0.237944	0.151188	0.221407	0.715093	0.741923
Solar Rad (W*m-2)	-0.18738	0.427317	0.339959	0.413051	0.516155	0.58142
Wind Direction	-0.25125	-0.14883	-0.21589	-0.16589	-0.07989	-0.01563
Wind speed	-0.55854	0.007221	-0.19902	-0.03942	-0.31078	-0.27816

	Solar Rad (W*m-2)						Wind Direction
	temp low	RH (%)	precip (in)	precip (cm)	UV-index		
precursor							
[M-H]- MW							
temp low	1						
	0.05805						
RH (%)	7	1					
	0.26053	0.35895					
precip (in)	8	2	1				
	0.26669	0.36336					
precip (cm)	3	9	1	1			
	0.62586	-	-	-			
UV-index	7	0.39869	0.38104	0.37176	1		
Solar Rad (W*m-2)	0.37508	-	-	-	0.73094		
	1	0.16741	0.46314	0.46392	5	1	
	-	-	0.02276	0.00590	-	0.18518	
Wind Direction	0.14606	0.13565	6	4	0.07066	6	1
	-	-	-	-	0.11509	0.07702	0.26621
Wind speed	0.37912	0.18345	0.25433	0.24658	3	6	9

Appendix 2: Observed OS and Average Concentrations

Suggested Formula	[M-H] ⁻ MW	Average concentration ng/m ³					Precursor Group	RT (min)
		August* (n=2)	Autumn (n=11)	Winter (n=7)	Spring (n=8)	June* (n=1)		
C ₃ H ₅ O ₅ S ⁻	152.99	0.8 (0.4)	0.7 (0.6)	0.3 (0.1)	0.8 (0.7)	0	Isoprene/ Anthropogenic ^{a-c,k,m}	1.21 _G
C ₃ H ₅ O ₆ S ⁻	168.98	0.6 (0.5)	0.7 (0.5)	0.6 (0.4)	0.8 (0.5)	0	Isoprene/Hexenal	1.12 _G
	168.98	0.3 (0.1)	0.3 (0.2)	0.2 (0.1)	0.5 (0.3)	0	Anthropogenic ^{a,b,d,i,k,m}	1.80 _G
C ₄ H ₇ O ₆ S ⁻	182.99	1.4 (1)	0.4 (0.5)	0.11 (0.08)	0.4 (0.7)	0	Isoprene/	1.13 _G
	182.99	0.7 (0.3)	0.3 (0.5)	0.08 (0.06)	0.4 (0.5)	0	Anthropogenic ^{b,e,f,m}	1.80 _G
C ₅ H ₇ O ₆ S ⁻	194.99	0.14 (0.1)	0.01(0.02)	0	0.04 (0.7)	0	Isoprene ^{i,m}	2.36 _G
C ₅ H ₉ O ₆ S ⁻	197.01	0.03 (0.02)	0.01 (0.01)	0	0	0	Isoprene ^{e,k,m}	2.69 _P
C ₄ H ₇ O ₇ S ⁻	198.99	3.16 (2.1)	0.9 (0.9)	0.12 (0.9)	0.8 (0.9)	0	Isoprene ^{a-c,k,m}	1.15 _G
	198.99	1.27 (0.7)	0.6 (0.7)	0.06 (0.04)	0.4 (0.4)	0		1.80 _G
C ₇ H ₁₁ O ₅ S ⁻	207.03	0.02 (0.02)	0.08 (0.01)	0.03 (0.04)	0.07 (0.9)	0.18	m	6.95 _{PHS}
C ₈ H ₁₅ O ₄ S ⁻	207.07	0	0	0.01 (0.01)	0	0	m	10.25 _{Oct}
C ₇ H ₁₃ O ₅ S ⁻	209.05	0.04 (0.01)	0.13 (0.1)	0.2 (0.1)	0.10 (0.07)	0.12	Dodecane ^{g,i,m}	7.16 _{PHS}
	209.05	0.03 (0.0)	0.3 (0.4)	0.4 (0.4)	0.1 (0.1)	0.05		7.36 _{PHS}
C ₆ H ₁₁ O ₆ S ⁻	211.03	0.2 (0.06)	0.5 (0.6)	0.4 (0.3)	0.4 (0.3)	0.78		6.03 _{PHS}
C ₇ H ₁₅ O ₅ S ⁻	211.06	0.01 (0.01)	0.06 (0.06)	0.12 (0.07)	0.05 (0.1)	0.04		7.82 _{PHS}
C ₅ H ₉ O ₇ S ⁻	213.01	2.0 (1.8)	0.5 (0.5)	0.15 (0.08)	0.6 (0.9)	0.77	Isoprene ^{b,d,e,m}	1.49 _G
	213.01	1.1 (0.7)	0.5 (0.7)	0.09 (0.04)	0.6 (0.8)	0.85		1.80 _G
	213.01	0	3.7 (11)	0	3.3 (6.5)	1.17		1.49 _G
C ₅ H ₁₁ O ₇ S ⁻	215.02	14.1 (13)	0.4 (0.7)	0	0	0	Isoprene ^{a-c,e,i,m}	1.33 _G
	215.02	4.2 (1.9)	3.2 (0.7)	0	1.6 (2.7)	0.74		1.76 _G
C ₇ H ₁₁ O ₆ S ⁻	223.03	0.14 (0.03)	0.09 (0.04)	0.09 (0.01)	0.2 (0.3)	0.72	Monoterpenes ^{b,i,k,m}	5.72 _{PHS}
C ₆ H ₉ O ₇ S ⁻	225.01	0.1 (0.1)	0.20 (0.04)	0.05 (0.05)	0.12 (0.08)	0.24	Hexenal/2-MeNAP ^{b,k,m}	3.00 _P
C ₉ H ₁₅ O ₅ S ⁻	235.06	0.13 (0.08)	0.9 (0.9)	0.5 (0.5)	0.6 (0.4)	1.3		7.89 _{PHS}
C ₈ H ₉ O ₇ S ⁻	237.01	0	0	0	0.01 (0.01)	0.02	Isoprene ^{b,i-m}	3.24 _P
C ₉ H ₁₇ O ₅ S ⁻	237.08	0	0	0.01 (0.01)	0	0	Dodecane ^{g,i-m}	9.61 _{Oct}
C ₇ H ₁₁ O ₇ S ⁻	239.02	0.2 (0.3)	0.10 (0.18)	0	0.2 (0.2)	0	Limonene ^{b,i-m}	4.48 _{PHS}
	239.02	0.12 (0.1)	0.15 (0.18)	0.01 (0.01)	0.2 (0.2)	0.06		4.82 _{PHS}
	239.02	0.08 (0.05)	0.2 (0.3)	0.2 (0.2)	0.3 (0.4)	1.18		5.42 _{PHS}
C ₉ H ₁₅ O ₆ S ⁻	251.06	0.09 (0.01)	0.4 (0.5)	0.2 (0.2)	0.2 (0.1)	0.36	Limonene ^{b,i,k,m}	7.61 _{PHS}
C ₆ H ₁₃ NO ₆ S ⁻	258.03	0	0.04 (0.1)	0.02 (0.05)	0	0	e	6.49 _{PHS}
C ₈ H ₁₁ O ₈ S ⁻	267.02	0	0	0	0.02 (0.03)	0		1.61 _{PHS}
C ₉ H ₁₅ O ₇ S ⁻	267.05	0.06 (0.09)	0.4 (0.6)	0.09 (0.6)	0.3 (0.2)	0.34	Isoprene/Limonene	7.16 _{PHS}
C ₁₀ H ₁₅ O ₇ S ⁻	279.05	0.11 (0.04)	0.42 (0.4)	0.2 (0.1)	0.5 (0.6)	0.48	Monoterpenes/ Anthropogenic ^{b,e,f,g,i-m}	6.21 _{Oct}
							Dodecane ^{g,i,k,m}	12.12 _{Oct}
C ₁₂ H ₂₃ O ₅ S ⁻	279.13	0	0.05 (0.07)	0.04 (0.05)	0	0		6.04 _{Oct}
C ₁₃ H ₁₄ O ₅ S ⁻	281.05	0	0	0	0	0		
C ₉ H ₁₅ O ₈ S ⁻	283.05	0.2 (0.2)	0.62 (0.5)	0.7 (0.4)	1.2 (0.7)	1.56	α-terpinene ^{b,i,k,m}	10.42 _{Oct}
C ₈ H ₁₄ NO ₆ S ⁻	284.04	0.02 (0.03)	0.08 (0.07)	0.11 (0.06)	0.2 (0.1)	0.23	m	10.42 _{Oct}
C ₁₄ H ₂₉ O ₄ S ⁻	293.18	0.05 (0.07)	0.2 (0.2)	0.3 (0.2)	0.14 (0.15)	0.34	k,m	14.08 _{Oct}
C ₁₀ H ₁₆ NO ₇ S ⁻	294.06	0.07 (0.09)	1.8 (1.7)	2.0 (2.0)	0.9 (0.5)	1.48	Monoterpenes ^{b,i,k,m}	10.86 _{Oct}
C ₉ H ₁₄ NO ₆ S ⁻	296.04	0.08 (0.1)	0.29 (0.3)	0.10 (0.06)	0.2 (0.1)	0.19	Limonene ^{b,i,m}	9.44 _{Oct}
C ₁₀ H ₁₇ O ₈ S ⁻	297.06	0.07 (0.7)	0.07 (0.1)	0	0.09	0.03	Monoterpenes/ Anthropogenic ^{b,e,g,i,k,m}	6.95 _{PHS}
							k,m	10.01 _{Oct}
C ₁₃ H ₂₅ O ₆ S ⁻	309.14	0	0	0	0	0		
C ₁₅ H ₂₉ O ₅ S ⁻	321.17	0	0.01 (0.01)	0	0.01 (0.01)	0.02	k,m	14.46 _{Oct}
C ₁₀ H ₁₆ NO ₉ S ⁻	326.05	0.04 (0.06)	0.18 (0.3)	0.06 (0.06)	0.2 (0.2)	0.3	Monoterpenes/ Anthropogenic ^{b,e,g,i,k,m}	7.61 _{PHS}
	326.05	0.01 (0.01)	0.09 (0.09)	0.05 (0.06)	0.04 (0.04)	0.05		7.91 _{PHS}
	326.05	0.4 (0.1)	0.14 (0.09)	0.1 (0.1)	0.13 (0.2)	0.4		7.87 _{PHS}
C ₁₀ H ₁₆ NO ₁₀ S ⁻	342.05	0.12 (0.06)	0.8 (0.2)	0.2 (0.2)	0.7 (0.9)	0.25	Monoterpenes ^{b,i,k,m}	8.16 _{PHS}
	342.05	0.01 (0.06)	0.3 (0.3)	0.06 (0.07)	0.2 (0.2)	0.03		9.16 _{PHS}
	342.05	0.11 (0.02)	0.3 (0.3)	0.06 (0.05)	0.4 (0.4)	0.29		10.30 _{Oct}
C ₁₆ H ₂₉ O ₇ S ⁻	365.16	0.01	0.01 (0.04)	0	0.02 (0.03)	0.05	k,m	14.08 _{Oct}
C ₁₀ H ₁₇ N ₂ O ₁₁ S ⁻	373.06	0.11	0.2 (0.4)	0.05 (0.03)	0.13 (0.2)	0.01	Limonene ^{b,i,k,m}	10.47 _{Oct}
C ₁₅ H ₁₆ O ₁₀ S ⁻	387.04	0	0.2 (0.3)	0.02 (0.06)	0.2 (0.2)	0.12	b	11.11 _{Oct}
C ₂₅ H ₂₂ O ₇ S ⁻	465.1	0.4 (0.3)	0.8 (2.1)	0.01 (0.02)	0.8 (0.7)	1.64		7.46 _{PHS}
C ₂₂ H ₁₃ NO ₉ S ⁻	466.03	0	0.01 (0.02)	0	0.02 (0.04)	0	b	11.34 _{Oct}
C ₃₉ H ₅₈ O ₄ S ⁻	621.39	0.13 (0.12)	0.07 (0.08)	0.3 (0.3)	0.6 (0.8)	0.22		11.75 _{Oct}

Months marked with an asterisk (*) had limited sampling days. Two sampling days occurred in August (8/29 and 8/30), and only one sampling day occurred in June (6/07). The June 6th date marks the front end of a hurricane. The standard deviations of each observed OS are shown in the parenthesis. The sources for each compound are as follows: a-Surratt et al. (2007); b- Surratt et al. (2008); c-Schindelka et al. (2013); d-Shalamzari et al. (2015); e- Noziere et al. (2010); f-Riva et al. (2016a); g-Riva et al. (2016b); h-Shalamzari et al. (2014); i-Riva et al. (2015).; j-Hansen et al. (2014); k-Tao et al. (2014); m-Kuang et al. (2015). References a-h are laboratory studies; i-m are field studies.

The surrogate used to quantify each compound is denoted as a subscript with the retention time. Surrogates were chosen based on similarity of retention time and structure. G=glycolic sulfate ester, L=lactic sulfate ester, P= propyl sulfate, Oct=octyl sulfate and PHS= 3-pinanol-2-hydrogen sulfate.

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