Evolution of Aerosol Under Moist and Fog Conditions in a Rural Forest Environment: Insights From High-Resolution Aerosol Mass Spectrometry

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Abstract The effect of foggy/moist conditions on the characteristics of secondary organic aerosol (SOA) was studied. Analysis of the high-resolution time-of-flight aerosol mass spectrometer measurements identified two fresh biogenic SOA factors (BSOA: BSOA-1 and BSOA-2), hypothesized to be formed through the nighttime reaction of biogenic volatile organic compounds with nitrate radical. During foggy periods, the more oxidized oxygenated OA (MO-OOA) was scavenged most efficiently (86% removal), followed by the less oxidized OOA (LO-OOA) (55%), SO4 (53%), and BSOA (25%). Aerosol mass increased during fog events, and the post-fog aerosol mass was higher (11%) than during the prefog period, with a reduction for MO-OOA and an enhancement for LO-OOA and BSOA. The high positive correlation observed between aerosol liquid water (ALW) and BSOA suggests the importance of aqueous-phase processing on the BSOA formation, and an increase in log10(ALW) of 1 corresponded to an average increase of about 0.9 µg m−3 in BSOA.

Plain Language Summary Ground-level aerosols impact human health, climate, and ecosystem health. Aerosols have complex chemical composition, and the chemistry of the carbonaceous fraction is a topic of great interest to atmospheric chemistry and air quality. Aerosol liquid water is an important component of these particles in humid environments, and it becomes the dominant component if the particle grows to become a fog or cloud droplet. In addition to influencing particle growth, the aerosol liquid water provides another medium in which chemical reactions can occur and produces species that remain in the condensed phase and thereby increase aerosol mass loading. This study investigated the evolution of aerosol mass and chemical composition in moist and foggy nighttime environments in a rural forested area of the Northeast U.S. Organic aerosols that were identified as biogenic secondary organic aerosols were observed to increase in mass concentration during foggy conditions, and in this environment, they increased linearly in mass as a function of the base 10 logarithm of aerosol liquid water over a wide range. The influence of aerosol liquid water on the chemical reactivity of aerosols could help to bridge the gap between modeled and observed secondary organic aerosol mass.

1. Introduction

Mass concentration and composition of atmospheric aerosols are important due to their influences on the radiation budget and climate, the formation of clouds, air quality, and human health (Biswas & Wu, 2005; McMurry, 2000). Among ambient aerosols, secondary organic aerosols (SOAs) are major constituents and generally formed through gas-to-particle conversion (Hallquist et al., 2009). Despite recent advances, there still exists great uncertainty in the understanding of the detailed formation of SOA, which may involve reactions of thousands of chemical species via different formation pathways, including new particle formation, condensation, and multiphase chemical reactions, particularly aqueous phase reactions (Ervens et al., 2011; McNeill, 2015).

Biogenic volatile organic compounds (BVOCs) are emitted from vegetation, particularly mature forests, then undergo atmospheric oxidation, and sometimes condense onto preexisting particles to form biogenic SOA.
(BSOA) (Lee et al., 2016), a major component of SOA in many environments. Advanced aerosol mass spectrometry technologies have been widely used for identifying newly formed BSOAs through their mass spectral fragments. For example, the $m/z$ 82 (C$_5$H$_6$O$^+$) fragment from the high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) mass spectrum has been used as a tracer for isoprene epoxydiol-derived SOA (Hu et al., 2015). Many AMS studies have been carried out in the southern United States (Ng et al., 2017; Xu et al., 2015, 2018), while there are fewer observations in the Northern United States and Canadian regions (Lee et al., 2016), which in general have different BVOC species, temperatures, solar radiation (SR) conditions, relative humidity (RH), and oxidant gas concentrations, and so on. Previous AMS measurements taken during a summer period in British Columbia, Canada, showed no significant ion signal at $m/z$ 82 but a high ion signal at $m/z$ 91 (C$_7$H$_7^+$) (Lee et al., 2016). Similarly, enhanced ion signal at $m/z$ 91 was also observed in the mass spectra of SOA formed from plant chamber experiments (Boyd et al., 2015; Kiendler-Scharr et al., 2009). To better understand the formation of BSOA in northern regions, more in-depth investigations are still needed.

Under conditions with high humidity, hygroscopic aerosols absorb water vapor to form aerosol liquid water (ALW), which is an abundant medium that facilitates partitioning of polar, water-soluble organic gases to the condensed phase, and can enhance SOA formation through oligomerization and functionalization of these organics via aqueous-phase oxidation (Lee et al., 2012). A recent study in a Japanese suburban forested area suggested that ALW can significantly promote water-soluble organic nitrogen (WSON) formation in submicrometer aerosol particles at this site (Xu et al., 2020). Under supersaturated conditions, such aerosols continue growing through additional water vapor condensation and can be activated to form fog or cloud droplets (Gilardoni et al., 2014). In the case of fog formation, fog drops also scavenge some fraction of the unactivated interstitial aerosols, and the compounds in fog drops may undergo aqueous reaction or be removed through direct droplet deposition (i.e., impaction or sedimentation) (Mandariya et al., 2019).

Many previous studies have examined fog processing in urban areas (Chakraborty et al., 2016; Ge et al., 2012; Herckes et al., 2013, 2015; Kim et al., 2019), where they are greatly affected by anthropogenic emissions, while the influence of fog processing on the BSOA, especially its aqueous processing mechanisms, is still poorly understood. Here we provide direct observations from a northeast U.S. rural forested area that examined the influence of fog processing on BSOA formation using an Aerodyne HR-ToF-AMS and additional instruments. This study presents a detailed investigation of fog processing focusing on (1) the SOA source apportionment; (2) the influence of different fog event phases or periods during the fog events on the aerosol composition and oxidation state; and (3) the relationship between ALW and BSOA formation.

### 2. Methods

#### 2.1. Sampling Location and Periods

The aerosols were sampled at Pinnacle State Park (henceforth PSP, latitude: 42.09°N, longitude: 77.21°W, elevation: 504 m) in Addison, New York, which is surrounded by mixed deciduous and coniferous forests. The village of Addison (population 1,800) is about 4 km to the northeast, and the city of Corning (population 12,000) is 15 km to the northeast (Schwab et al., 2009). In this study, we focus primarily on the measurements from 19–23 September 2017, during which several fog events occurred. To provide a contrast to the foggy nighttime periods, a 1-day measurement period from 11 to 12 April 2018 was used to explore the influence of temperature on the aerosol evolution during overnight measurement periods (Text S1).

#### 2.2. Aerosol Measurements

Submicron aerosols were measured at PSP using a mobile measurement platform (a 2007 Dodge Sprinter van), including an HR-ToF-AMS and a TSI Scanning Mobility Particle Sizer Spectrometer (SMPS) (Zhang et al., 2019, 2020). The HR-ToF-AMS was used for measuring the chemical composition of the non-refractory (NR) submicron aerosol (NR-PM$_2.5$) (DeCarlo et al., 2006; Drewnick et al., 2005), including organics, particle-sulfate (SO$_4$), particle-nitrate (NO$_3$), particle-ammonium (NH$_4$), and particle-chloride (Cl). During the PSP measurements, the HR-ToF-AMS was operated in “V” mode with an averaging period of 5 min.

In addition, several trace gas analyzers (Table S1 in the supporting information) were on site to measure trace gases, including O$_3$, NO, NO$_y$, CO, SO$_2$, and HNO$_3$; meteorological sensors provided temperature,
RH, SR, wind speed, and wind direction. For consistency, the final average time resolution reported and analyzed for all continuous instruments was set to 5 min. In the mobile laboratory, ambient air at a flow rate of approximately 56 L min$^{-1}$ was drawn into a 2.5 cm outside-diameter stainless steel inlet tube, which connected downstream to a PM cyclone (URG-2000-30EC), which removed particles and droplets larger than about 2 $\mu$m. The AMS and SMPS flows were taken from a roughly 3 L min$^{-1}$ subsample of the main aerosol flow that first passed through a silica gel diffusion dryer, which reduced the RH of the subsample to less than 40% in order to reduce uncertainties in the bounce-related collection efficiency (CE) and possible losses in particle transmission due to the particle hygroscopic growth under high RH conditions (http://cires1.colorado.edu/jimenez-group/wiki/index.php/Field_ToF-AMS_Operation). While it is also possible that some semi-volatile species (in addition to water) may also be lost during the drying process, this effect appears to be small according to a study on fog water chemistry conducted in Central Valley, California, where a comparison was made between residual aerosol composition measured by HR-ToF-AMS versus fog water composition measured by ion chromatography (IC) and total organic carbon (TOC) analysis (Kim et al., 2019). The cutoff of the PM cyclone and of the instruments themselves did not allow direct measurement of most fog drops, but some fine fog drops could still pass through the PM cyclone and evaporate in the silica gel dryer to form residual aerosols. During foggy periods, the instruments measured both the unactivated interstitial aerosols and the residual aerosols from the fine fog drops that evaporated in the silica dryer. For simplicity, these forms of aerosol are collectively defined as interstitial aerosols in this study.

3. Data Analysis

The HR-ToF-AMS data were analyzed using the standard HR-AMS data analysis software, SQUIRREL v1.59D and PIKA v1.19 (Allan et al., 2003; Canagaratna et al., 2007). During the analysis, the default relative ionization efficiency (RIE) values were used. The applied CE of 0.5 followed a suggested value used in previous studies (Zhang et al., 2005). The PMF Evaluation Tool (PET, v2.08D) was used to divide the organic matter into different organic factors based on the PMF2 algorithm (Ulbrich et al., 2009), and the final result was verified based on the criterion method demonstrated in previous studies (Zhang et al., 2011). The elemental analysis (oxygen-to-carbon [O:C] and hydrogen-to-carbon [H:C] ratio) is calculated based on the improved method proposed by Canagaratna et al. (2015).

The ALW was estimated as a sum of calculated contributions from the aerosol inorganic and organic components determined by the AMS. The ALW contributed by inorganic aerosol compounds was calculated using the thermodynamic model ISORROPIA-II (Ding et al., 2019; Fountoukis & Nenes, 2007), which uses the HR-ToF-AMS particle mass concentrations of the inorganic species (SO$_4$, NO$_3$, NH$_4$, and Cl), the ambient RH, and ambient air temperature as the inputs (Text S2). For the calculation of ALW from the organic aerosol compounds, an empirical equation was used (Nguyen et al., 2016; Xu et al., 2020):

$$\text{ALW} = \frac{M_{\text{org}}}{\rho_{\text{org}}} \times x_{\text{org}} \times \frac{a_w}{1-a_w}$$

Here, $M_{\text{org}}$ is the mass concentration of organic compounds, $\rho_{\text{org}}$ is the density of organic compounds (1.4 g cm$^{-3}$ is used; Nguyen et al., 2016; Xu et al., 2020), $x_{\text{org}}$ is related to the organic hygroscopicity (0.1 is used; Nguyen et al., 2016; Xu et al., 2020), and $a_w$ is the water activity ($a_w$ is taken to be equal to RH; Nguyen et al., 2016; Xu et al., 2020).

4. Results and Discussion

Figure S1 shows the time series of aerosol components, meteorological parameters, and trace gases from 19–23 September 2017. During the measurement period, the dry aerosol PM$_1$ mass concentration at PSP was 1.3–8.5 $\mu$g m$^{-3}$ with an averaged value of 5.6 $\mu$g m$^{-3}$. Organics were the main component of PM$_1$, and, on average, comprised about 76% of the aerosol mass concentration, with proportions as high as 84%. The average mass fractions for other species were 14% for SO$_4$, 2% for NO$_3$, 7% for NH$_4$, and 1% for Cl. During this period, there were four nights with maximum RH > 96%, and the first three nights were verified as having fog conditions from images captured by the camera from the Addison New York State Mesonet.
station (Text S1 and Figure S2; Brotzge et al., 2020). At the same time, during the four overnight periods, the temperature was generally below 15°C, and the wind speed was generally below 1 m s⁻¹ from the west, indicating a stagnant boundary layer with cool temperatures and moist conditions (Figure S1).

4.1. OA Source Apportionment

Four OA factors, including one more-oxidized oxygenated OA (MO-OOA), one less-oxidized oxygenated OA (LO-OOA), and two biogenic secondary OAs (BSOAs: BSOA-1 and BSOA-2), were identified by the PMF analysis. Detailed information of the PMF results is shown in the supporting information (Text S3 and Figures S4–S7). Briefly, following previous studies (Şeytan et al., 2012; Wu et al., 2019; Xu et al., 2015, 2018), we named the factor with the highest O:C ratio (0.86) as MO-OOA and the factor with the second highest O:C (0.76) as LO-OOA. The MO-OOA represented a highly oxidized and more aged organic aerosol due to its highest degree of oxygenation and showed a daily maximum during the late afternoon or early evening (around 19:00 local time, Figure S5c). The LO-OOA represented a fresher organic aerosol with a daily maximum at early afternoon (around 14:00 local time, Figure S5c). Meanwhile, LO-OOA showed variation that correlates better with SO₄ (R² = 0.36) than with NO₃ (R² = 0.02), suggesting similar oxidative processing for LO-OOA and sulfate, possibly through OH oxidation given the similar maxima times of the LO-OOA and solar irradiation time series (Figure S8a). In contrast, the peak periods of MO-OOA matched more closely the O₃ or O₃ variation, especially for the first 3 days, consistent with its higher oxidation state (Figure S8b).

The two BSOAs were fresher with lower O:C ratio than MO-OOA and LO-OOA, and their mass spectra profiles featured a high signal at m/z 91 (C₆H₇⁺, average f₉₁ of 0.01, Figure S5a), which has been shown in previous work to be produced through the reaction of BVOC with nitrate radical (Boyd et al., 2015; Kiendler-Scharr et al., 2009; Takeuchi & Ng, 2019). The least oxidized factor is designated as BSOA-1 and the other as BSOA-2. Both BSOA-1 and BSOA-2 exhibited their highest concentrations during the night (Figure S5c) and decreased during the afternoon. The sum of BSOA-1 and BSOA-2 showed high correlation with organic nitrate (hereafter NO₃,org, Text S4) with an R² of 0.64 (Figures S9a and S9c), lending credibility to the hypothesis that BSOA and NO₃,org formed through nighttime nitrate radical (NO₃) + BVOC reactions (Boyd et al., 2015, 2017; Farmer et al., 2010; Fry et al., 2009; Rollins et al., 2012). The calculated NO₃,org time series also showed correspondence with NO₃,ion and HNO₃ (Figure S9), indicating the synchronous formation pathway for NO₃,org and NO₃,ion during the nighttime. HNO₃ nighttime production could be due to the reaction of gas-phase nitrate radical with hydrocarbons and/or the hydrolysis and evaporation of the formed NO₃,org and NO₃,ion (Ng et al., 2017; Takeuchi & Ng, 2019). There is evidence from the aerosol mass spectra that BVOCs were monoterpenes (most likely β-pinene) due to the enhanced organic nitrate fragments signals at m/z 45 (CH₃NO⁺) and m/z 76 (CH₂NO₃⁺) (Figure S9b) with elevated BSOA and NO₃,org concentration. The m/z 45 and m/z 76 signals were shown to be proxies for the production of nitrate radical + β-pinene in a chamber study (Bruns et al., 2010). Similar to observations of Bruns et al. (2010), the intensity of these fragment signals is quite weak, though they were unique to organic nitrates.

BSOA-1 shows a higher H:C ratio than BSOA-2, while BSOA-2 has the similar mass spectra as LO-OOA (R² = 0.96, p < 0.01), with lower O:C (0.59 vs. 0.78) and OM:OC (1.92 vs. 2.14) ratios but a similar H:C ratio (1.39 vs. 1.42) and similar f₄₄ (0.138 vs. 0.144), likely indicating the addition of OH/OOH functional groups on the carbon skeleton through OH oxidation (He et al., 2018). One plausible mechanism to explain this is that BSOA-1 was the initial product of the nitrate radical + BVOC reaction during nighttime, and then BSOA-1 was fragmented and/or oxidized further to form BSOA-2, which converted to LO-OOA via photo-oxidation reactions (OH reaction) and further oxidized to form MO-OOA (Ng et al., 2017). Given that these two BSOAs are closely related based on their diurnal variation and mass spectral characteristics, they are considered together (BSOA = BSOA-1 + BSOA-2) in the following sections.

4.2. Fog Events

Three fog episodes were observed (the nights of 19–20, 20–21, and 21–22 September) during the study period, while the fourth night (22–23 September) was characterized by high moist conditions with high RH (RH > 96%) but no visually discernable fog. Based on the transition point times of the aerosol compounds (that is, a noticeable step change in concentration), as well as RH and solar irradiation variations, the entire fog event life cycle was divided into five fog processing phases or periods in a manner similar to a previous study (Mandariya et al., 2019), namely, fog_before_forming (F-B), fog_forming (F-F), fog_scavenging (F-S), fog_ending (F-E), and fog_withdrawal (F-W).
(F-S), fog_dissipating (F-D), and fog_post_dissipating (F-P). The detailed properties for each fog phase can be found in the supporting information (Text S4 and Table S2). There were no F-S and F-D periods during the night of 22–23 September, mostly due to the lower peak RH values and the absence of fog (Figures S2d and S10). The F-S periods of 19–20 and 20–21 September were further divided into another two subperiods labeled F-S-L for light fog conditions and F-S-H for heavier fog conditions based on the visibility variation, as shown in Figures S10 and S11 and Table S2. During the fog scavenging periods for each day, as shown in Figures 1 and S10, most of the aerosol compound mass concentrations experienced significant reductions. We define the scavenging efficiency (Table S3) that occurred during these periods as (Mandariya et al., 2019)

$$\text{SE} = \frac{C_{F-B} - C_{F-S}}{C_{F-B}} \times 100\%$$

where SE is the scavenging efficiency and C is the mass concentration for all aerosol compounds except BSOA and NO₃. For BSOA, $C_{F-F}$ was used to replace $C_{F-B}$ due to their enhancement during the F-F period.

The observed scavenging was most dramatic during these fog events for MO-OOA and occurred during F-S-H periods, indicating its highest solubility and scavenging efficiency (SE_MO-OOA ranged from 77–92% with an averaged value of 86%, Table S3). LO-OOA and SO₄²⁻ were the other two species that were readily scavenged, with fairly high and similar averaged scavenging efficiencies (SE_LO-OOA: 30–86% with an averaged value of 55%, SE_SO₄²⁻: 40–73% with an averaged value of 53%, Table S3). BSOA increased during the F-F period compared to the F-B period, before decreasing during the F-S period. Consequently, BSOA showed lower scavenging efficiencies (16–36% with an averaged value of 25%, Table S3) compared to LO-OOA and SO₄²⁻, presumably due to the compensation of BSOA production during the preceding high RH conditions.

The high scavenging efficiency of LO-OOA and MO-OOA resulted in a decreasing O:C ratio during the fog forming and scavenging periods, and with the dissipation of fog, the mass concentration of the residual aerosol from the evaporation of the fog drops increased for the interstitial aerosol, accompanied by an increase in the O:C ratio (Figure 1). The aerosols during the F-P period are composed of both residual aerosols from fog drops and the interstitial aerosols remaining from the F-S period. Compared to the initial period (F-B), the
total mass concentrations of the F-P aerosols was higher by about 7% to 15% for the first three nights with an averaged value of 11%, and the O:C ratios decreased while the H:C ratios increased, which is likely related to the decrease of MO-OOA and the enhancement of LO-OOA + BSOA. For the organic factors, MO-OOA showed an averaged reduction of 0.71 μg m⁻³ (73% reduction), which was possibly due to fog droplet removal by sedimentation. However, this reduction was in contrast to the behavior of LO-OOA and BSOA, which showed clear averaged enhancements of 0.52 μg m⁻³ (33% enhancement) for LO-OOA and 0.58 μg m⁻³ (41% enhancement) for BSOA. When combining LO-OOA and BSOA together, the enhancement for each fog event was relatively consistent with 1.27, 1.07, and 0.95 μg m⁻³ enhancements for the nights of 19–20, 20–21, and 21–22 September, respectively (Figure 1).

In contrast to the first three nights, the night of 22–23 September lacked significant F-S and F-D periods. LO-OOA and MO-OOA continued decreasing during the high RH period (RH > 96%), while the BSOA kept increasing until 04:30 am (local time) with a near-constant rate of 0.32 μg m⁻³ hr⁻¹ (Figure S9), which is consistent with a calculation of BSOA formation from β-pinene and nitrate radical (NO₃) based on the two-product model (Griffin et al., 1999; Odum et al., 1996, 1997; Russell & Allen, 2005; Text S5). In the absence of fog on the night of 22–23 September, the total mass concentration during F-P was larger than during F-B, with reductions of 0.34 μg m⁻³ (65% reduction) for MO-OOA and 0.15 μg m⁻³ (14% reduction) for LO-OOA and an enhancement of 1.2 μg m⁻³ for BSOA (48% enhancement). Compared to the three foggy nights, the lower reduction of MO-OOA during the 22–23 September night reflected the weaker aerosol scavenging in nonfoggy, highly moist conditions (RH > 96%). There was some decrease for LO-OOA during the night of 22–23 September, which was different from the three foggy nights. However, the enhanced BSOA mass concentration during 22–23 September was close to the combined enhancement of LO-OOA and BSOA on the foggy nights. This indicates that (1) the enhancement of “(BSOA+LO-OOA) on the foggy nights” or “BSOA on the non-foggy night” is associated with high RH conditions rather than the existence of fog drops and (2) there may have been a transition from BSOA to LO-OOA in the fog drops on the three foggy nights. This study is limited by relatively few measurement days and instruments. To further understand the transition from BSOA to LO-OOA, more measurements with a fog droplet collection system, such as a counterflow virtual impactor (CVI), are required.

4.3. Observed Relationship Between BSOA and ALW

The low wind speed, high RH values, nighttime conditions (i.e., no shortwave radiative heating of the surface), and observed fog formation present a situation with weak vertical mixing, which rules out dynamics and transport as the dominant mechanism for changes in aerosol composition during foggy periods. The rural location is free of significant surface pollutant emissions, and the previous day’s residual mixed layer is not significantly different from the stable nocturnal boundary layer in pollutant concentrations. These dynamics provide a nearly ideal environment for investigating aerosol evolution and the effect of RH (or ALW) on the newly formed BSOA (BSOA-1 + BSOA-2). Figures 2a and S12a–S12c show the scatterplot of RH and the BSOA mass concentration for each night of the September 2017 period (only F-B and F-F periods are considered). Markers are colored by temperature and sized by calculated ALW values. As shown in the figures, when RH increased and was below 96%, BSOA mass concentrations also increased, consistent with the results from chamber and modeling studies (Pye et al., 2017; Stirnweis et al., 2017; Takeuchi & Ng, 2019). The increased RH (or ALW) (1) provides more medium for gas-soluble species to dissolve and also reduces the viscosity and increases the diffusivity of the quasi-aqueous organic reactant molecules, increasing the aerosol bulk reaction rate to produce more BSOA (Shiraiwa et al., 2011; Takeuchi & Ng, 2019), and/or (2) reduces the viscosity and increases the diffusivity of small size BSOA aerosol or related organic matter (such as highly oxygenated organic molecules [HOMs]), which would be more easily or quickly dissolved into ALW and mixed with other inorganic/organic components and enhance the particle mass/size growth, which in turn are measured by the HR-ToF-AMS (Shiraiwa et al., 2017; Slade et al., 2019). This is partly supported by the good agreement between BSOA mass concentration with the SMPS measured aerosol number concentration in Aitken mode (60–110 nm, Figure S13), and this mode was near the lower cut off size of the HR-ToF-AMS aerodynamic lens (70 nm for 100% transmission efficiency). To better understand the effect of ALW on the BSOA mass concentration, the relationship between these two parameters is shown in Figure 2b. The linear regression analysis between the BSOA mass concentration and log₁₀(ALW) for each
night showed moderate to high correlation coefficients ($R^2 = 0.70$–$0.96$, $p < 0.01$). The slopes of these four nights are $0.6$–$1.3$ with an averaged value of $0.9$, which implies during this study that an increase of $1$ in the $\log_{10}(ALW)$ would lead to an increase of about $0.9 \, \mu g \, m^{-3}$ BSOA (on average).

During the periods of increasing RH, the air temperature decreased, which tended to keep the BSOA stable in the aerosol phase and enhance the organic compound concentration (Lee et al., 2014). However, the decreased temperature could also suppress the BSOA formation through several possible ways: (a) lower BVOC emissions from trees (Tarvainen et al., 2005); (b) reduce the reaction rates (Ervens et al., 2011; von Hessberg et al., 2009); and (c) reduce the diffusion coefficients and increase the viscosity of organic molecules or small size organic matter (Kiland et al., 2019; Price et al., 2015). Chamber studies show a positive relationship between SOA yields and temperature under high humidity conditions (von Hessberg et al., 2009). The single day of measurements in 2018 occurred in much colder overnight temperatures (2–6°C) with high RH (90–95%) but no fog formation (Text S1). BSOA concentration did increase during this 2018 overnight period (Figure S12d) with a maximum slope (BSOA vs. $\log_{10}(ALW)$) of $0.29 \, \mu g \, m^{-3}$ (Figure 2b), values significantly less than the slopes between $0.56$ and $1.33 \, \mu g \, m^{-3}$ for the 2017 overnight periods. This indicates that high RH conditions (and the associated ALW) was more effective at promoting BSOA mass enhancement rather than lower temperatures in this study. Further lab chamber experiments in high RH conditions (from about 60% to 95%) over a range of temperature, as well as model simulations, are still warranted to better describe the nighttime BSOA formation under conditions of high RH. Nonetheless, our fitting results between BSOA and ALW may give an estimation of BSOA formation in this rural forested area of the northeastern USA under similar mass loading, RH/ALW and temperature conditions, which are common during late summer and early fall (as shown in Figure S14). These observations and results should help improve the estimation of the organic aerosol mass formation in atmospheric models and encourage additional work in the area of aqueous processing of aerosols.

5. Conclusion

In this study, the organic aerosol composition at Pinnacle State Park, NY, including the BSOA hypothesized to have formed through nitrate radical + BVOC reaction, was measured using an Aerodyne HR-ToF-AMS and collocated instruments. The effect of foggy/moist conditions and fog processing on the characteristics of secondary aerosol was studied with special emphasis on scavenging efficiency and the aerosol comparison between different phases of the fog events. A high positive correlation existed between RH/or ALW and BSOA, which could be used to help improve the estimation of the organic aerosol mass formation during cool and moist nighttime conditions in Northeast forested regions.
Conflict of Interest
The authors declare that they have no conflict of interest.

Data Availability Statement
The data used in this manuscript are listed in the references, tables, and supplementary materials and are accessible at http://atmoschem.esrc.cesm.albany.edu/~aqm/AQM_Products/paper_datasets/.

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