Cloud, Aerosol, and Radiative Properties over the Western North Atlantic Ocean

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Key Points:
- Both surface radiation and reanalysis aerosol data show similar differences between weather states (WSs) at two sites in the study region
- The clear sky WS exhibited below average seasonal values of aerosol optical thickness (AOT) at both sites
- Analysis of WS transitions showed that more convective WSs were more prevalent as ending states under high AOT conditions
Abstract
This study examines the atmospheric properties of weather states (WSs) derived from the International Satellite Cloud Climatology Project (ISCCP) over the Western North Atlantic Ocean. In particular, radiation and aerosol data corresponding to two sites in the study domain, Pennsylvania State University and Bermuda, were examined to characterize the atmospheric properties of the various satellite-derived WSs. At both sites, the fair weather WS was most prevalent, followed by the cirrus WS. Differences in the seasonality of the various WSs were observed at the two sites. Fractional sky cover and effective shortwave cloud transmissivity derived from ground-based radiation measurements were able to capture differences among the satellite-derived WSs. Speciated aerosol optical thicknesses (AOT) from the Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) were used to investigate potential differences in aerosol properties among the WSs. The clear sky WS exhibited below-average seasonal values of AOT at both sites year-round, as well as relatively high rates of occurrence with low AOT events. In addition, the clear sky WS showed above-average contributions from dust and black carbon to the total AOT year-round. Finally, transitions between various WSs were examined under low, high, and mid-range AOT conditions. The most common pathway was for the WSs to remain in the same state after a 3 hr interval. Some WSs, such as midlatitude storms, deep convection, middle top, and shallow cumulus, were more prevalent as ending states under high AOT conditions. This work motivates examining differences in aerosol properties between WSs in other regions.
1. Introduction

The Western North Atlantic Ocean (WNAO) represents a region of diverse meteorological and atmospheric conditions, due in part to pollution outflow from North America to the region (e.g. Anderson et al., 1993; Fehsenfeld et al., 2006; Li et al., 2005) and in part to the distinct oceanic and atmospheric circulations within the Atlantic Ocean basin (e.g., Davis et al., 1997; Hurrel, 1995). Because of the accessibility of the region and the wide range of conditions present, over 700 works have been published in the realm of atmospheric sciences for the WNAO and adjacent North American east coast (Sorooshian et al., 2020). However, a review of these works reveal that only 2% are focused on aerosol-cloud interactions (Sorooshian et al., 2020), which represent the largest uncertainty in the radiative forcing of climate (Myhre et al. 2013). Despite the deep body of research, questions still remain as to the interactions among aerosols, clouds, radiation, and meteorology in the WNAO and the significance of changes in this region over time to the global climate.

Clouds are the primary modulators of radiation at the Earth’s surface and affect large variations in surface irradiance over a range of time scales, from minutes to decades. Clouds are therefore principal in understanding changes in climate. While the radiative properties of clouds depend strongly on cloud type, the impact of aerosol on cloud properties also varies with cloud and meteorological regime. Aerosol indirect radiative effects are relatively strong in marine stratocumulus decks within the subtropical eastern ocean basins (e.g. Mulmenstadt et al. 2019) and have been widely studied for their response to aerosol (Sorooshian et al., 2019) due to the importance of the resulting radiative effects but also due to the fact that observations of key cloud properties and their variability with aerosol are more easily accessible in these relatively uniform cloud structures. Resolving the radiative impacts of aerosol in diverse cloud fields of complex morphology requires directed observational approaches. In a region such as the WNAO with a high diversity of both aerosol and meteorological conditions, a large observational set is required to separate drivers of variability in cloud properties and surface radiation.

In order to address some of the outstanding questions related to aerosol-cloud-precipitation-meteorology interactions for the WNAO, various multi-year in situ measurement campaigns have recently occurred or are currently in progress, including the North Atlantic Aerosols and Marine Ecosystems Study (NAAMES; Behrenfeld et al., 2019) and the Aerosol Cloud Meteorology Interactions over the Western Atlantic Experiment (ACTIVATE; Sorooshian et al., 2019). Analysis of aerosol-cloud-precipitation-meteorological interactions over long time frames can help contextualize results from shorter-term airborne missions. Both space- and ground-based observational records provide long-term, continuous data records of key aerosol, cloud and radiation parameters that can be used to define the co-variability of these parameters.

In a study using data from the National Oceanic and Atmospheric Administration (NOAA) Surface Radiation budget observing network (SURFRAD) across the United States, as well as the Department of Energy’s (DOE) Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) network, Long et al. (2009) found an increase in shortwave downwelling radiation (SWdn) from 1995-2007, which was corroborated in subsequent studies using SURFRAD data from 1996-2011 (Augustine and Dutton, 2013) and 1995-2010 (Gan et al., 2014). Long et al. (2009) and Augustine and Dutton (2013) contend that the documented decreases in aerosol optical depth (AOD) over the United States did not contribute a large amount to the observed increases in SWdn, but rather indicated that changes in cloudiness were the main factor. However, Gan et al. (2014) note that changes in AOD could impact cloudiness through aerosol
indirect effects. Using SURFRAD observations, Ten Hoeve and Augustine (2016) show a strong impact of aerosol on cloud cover, and thus surface irradiance, over the US.

Satellite-borne retrievals of cloud properties, including cloud-top pressure and optical depth, have been used to identify similar types of products that have been given different names, including global “cloud regimes” (Oreopoulos et al., 2014), “weather states” (WSs; Tselioudis et al., 2013) and “nodes” (McDonald et al., 2016), as well as “cloud types” on a regional scale (Hill et al., 2018). These cloud regimes and WSs have proven to be useful tools in constraining analyses of atmospheric regimes and evaluating climate models (Remillard & Tselioudis, 2015; Williams & Brooks, 2008; Williams & Webb, 2009). Datasets from the International Satellite Cloud Climatology Project (ISCCP) and the Moderate Resolution Imaging Spectroradiometer (MODIS) instruments onboard the National Aeronautics and Space Administration (NASA) Terra and Aqua satellites have been utilized many times to derive these WSs/cloud regimes and understand their distinctive radiative properties (Oreopoulos et al., 2016; Oreopoulos & Rossow, 2011). In addition, previous studies have highlighted the importance of constraining aerosol-cloud interaction analyses within distinct cloud regimes derived using MODIS data (Gryspeerdt & Stier, 2012; Oreopoulos et al., 2017; Oreopoulos et al., 2020). Therefore, the WSs can provide a backdrop against which to better quantify aerosol-cloud-radiation-meteorology interactions and evaluate long-term trends in atmospheric regimes.

Given the uncertainties in mechanisms contributing to long-term changes in the radiation budget, and specifically in the role of aerosol in forcing changes in cloud radiative properties across different cloud types and meteorological regimes, improved understanding of the impacts of aerosol-cloud-meteorology interactions on the radiation budget is necessary. This study seeks to present a link between ground-based measurements, satellite retrievals, and reanalysis results in order to provide context for ongoing and future measurements over the U.S. East Coast and WNAO. Furthermore, by characterizing the distinct radiative and potentially distinct aerosol environments of the various WSs, changes in WS frequency or spatial distribution can be used as a proxy to estimate the changes in other characteristics, such as radiative environment.

2. Methods

2.1 Weather States

Weather States (WS) are derived through the application of a clustering algorithm on Cloud Optical Thickness-Cloud Top Pressure (TAU-PC) histograms from the International Satellite Cloud Climatology dataset (Rossow and Schiffer 1991), which produces the dominant cloud structures that characterize cloud property variability. A global WS dataset was originally derived using the 2.5-degree resolution ISCCP-D dataset by Tselioudis et al. (2013), and this dataset was recently updated using the latest, 1-degree resolution, ISCCP-H dataset (Young et al., 2008) and is presented in Tselioudis et al. (2021). The WSs are produced at $1° \times 1°$ spatial resolution every 3 hours during daylight. Note that this introduces a variation in the number of states described per day given the time of year (e.g. more daylight hours in the summertime yields additional WSs per day). The latest ISCCP-H WS dataset includes 10 distinct cloud WSs plus one state of “clear sky.” The dataset together with the WS TAU-PC histograms and their geographical distributions can be found at https://isccp.giss.nasa.gov/wstates/hggws.html. As shown in Remillard and Tselioudis (2015) and in Tselioudis et al. (2021), the WS TAU-PC histograms correspond to cloud structures with well-known morphological characteristics, and Tselioudis et al. (2021) provide a naming convention for the ISCCP-H WSs after merging two WSs with similar TAU-PC histograms and geographical distributions. We follow this same
naming convention, and we present the WSs used in this paper in Table 1, following a progression from high-top to low-top WSs.

<table>
<thead>
<tr>
<th>WS Symbol</th>
<th>WS Description</th>
<th>WS Numbers from ISCCP-H Dataset</th>
</tr>
</thead>
<tbody>
<tr>
<td>DCN</td>
<td>Deep convection</td>
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</tr>
<tr>
<td>MDS</td>
<td>Midlatitude storms</td>
<td>2</td>
</tr>
<tr>
<td>CIR</td>
<td>Cirrus</td>
<td>3,6</td>
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<tr>
<td>PLR</td>
<td>Polar</td>
<td>4</td>
</tr>
<tr>
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<td>Middle top</td>
<td>5</td>
</tr>
<tr>
<td>FRW</td>
<td>Fair weather</td>
<td>7</td>
</tr>
<tr>
<td>SHC</td>
<td>Shallow cumulus</td>
<td>8</td>
</tr>
<tr>
<td>STC</td>
<td>Stratocumulus</td>
<td>9,10</td>
</tr>
<tr>
<td>CLR</td>
<td>Clear sky</td>
<td>11</td>
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</tbody>
</table>

2.2 Ground-based Radiation Measurements

Two sites bounding the study domain with long-term ground-based radiation measurements are the SURFRAD (Augustine et al., 2000) station at Pennsylvania State University (PSU) and the World Climate Research Programme (WCRP) Baseline Surface Radiation Network (BSRN; Ohmura et al., 1998) station at Bermuda (BER). These two sites coincidentally fall within the domain of interest (i.e. WNAO region) and are well-suited for better understanding aerosol-cloud-meteorology-radiation interactions in different aerosol regimes, as will be further discussed in Section 2.3.

Available data from these sites include relative humidity (RH) and results from the comprehensive Radiative Flux Analysis (RadFlux) developed by Dr. Charles N. Long, which uses broadband, hemispheric radiation data that have undergone quality assurance testing via the QCRad methodology (Long and Shi, 2006; 2008) in order to yield numerous derived quantities. Of particular interest to the present study are two quantities derived from measurements of total and diffuse shortwave downwelling radiation (SWdn), as calculated using algorithms from Long and Ackerman (2000) and Long et al. (2006): effective shortwave cloud transmissivity (defined as the ratio of SWdn to estimated clear-sky SWdn) and fractional sky cover (SWSev).

2.3 Aerosol Data

Surface-based retrievals of AOD are available from the SURFRAD site at PSU and from Aerosol Robotic Network (AERONET; Holben et al., 1998) sites located on the island of Bermuda. AOD data from PSU were not available for 2010-2012. Three AERONET sites have operated at Bermuda, with site names and years of operation as follows: Bermuda (1996-2002), Prospect Hill (2005-2006), and Tudor Hill (2007-2019). Level 2 AOD data from AERONET were downloaded when available in the range of 2000-2012 for these three sites at BER.

Due to cloud-interference and temporal gaps in instrumentation at both BER and PSU, aerosol data from a reanalysis model, the Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2), were used to provide improved temporal coverage. The MERRA-2 reanalysis provides total aerosol extinction (AOT) at 550 nm at 0.625° × 0.5° spatial resolution and time-averaged hourly temporal resolution (Randles et al., 2017). MERRA-2
assimilates bias-corrected remotely-sensed AOD when available (Randles et al., 2017). In addition, extinction AOT at 550 nm from MERRA-2 was analyzed for the following species: dust, sulfate (SO₄), sea salt (SS), black carbon (BC), and organic carbon (OC).

As a check to determine the validity of the MERRA-2 results at the two ground-based sample sites, Figure S1 in the supplement shows the comparison of the ground-based retrievals of AOD and MERRA-2 AOT results at 550 nm. Note that neither ground site provided AOD data at 550 nm, and thus the AOD at 550 nm was calculated based on the spectrum of AOD retrievals available at varying wavelengths using a second-order polynomial fitting equation (Eck et al., 1999). Values of AOD at 550 nm were only used if AOD data from at least three different wavelengths were reported and the resulting polynomial fit for the measured wavelengths had a coefficient of correlation ($R^2$) greater than 0.5. Furthermore, ground-based measurements were averaged to match the hourly temporal resolution of MERRA-2. For each site, a linear regression was performed using both a variable and fixed y-intercept (i.e. $y = 0$). Two instances of high AOD measured at BER from AERONET were removed before the linear regression was performed (AOD = 2.148, 6.216). The MERRA-2 results exhibited a stronger correlation with ground-based measurements at PSU versus BER (for linear regression with variable y-intercept, $R^2 = 0.71$ at PSU based on 7570 samples and $R^2 = 0.60$ at BER based on 7808 samples). For both sites, MERRA-2 under-predicts AOD relative to the ground-based retrievals.

While both sites lie within the WNAO region, they represent locations with distinct aerosol and atmospheric environments. The well-documented pollution outflow from the North American East Coast yields a gradient in aerosol from west to east in the region. The sites at PSU and BER bookend this domain such that PSU represents a continental site within the midst of the pollution sources, while BER is located at a more remote marine location heavily-influenced by long-range transport of aerosol (e.g. Dickerson et al., 1995; Keene et al., 2014). The locations of the two sites, as well as seasonal averages (December-January-February (DJF), March-April-May (MAM), June-July-August (JJA), September-October-November (SON)) of AOT at 550 nm from MERRA-2 can be found in Figure 1.
Figure 1. Seasonal averages of AOT (550 nm) from MERRA-2 for the study region based on data between 2000 and 2012. The locations of the ground-based measurement sites at Pennsylvania State University (PSU) and Bermuda (BER) are also indicated. Hourly data with nominal times from 14:30 – 18:30 GMT daily were used to produce the figure. These times correspond to daylight hours with WS data available year-round.

As the hourly-averaged MERRA-2 AOT data are centered at 30 min after the hour (e.g. 02:00 – 03:00 corresponds to the AOT at the nominal time of 02:30) and the WS data are centered on the hour every three hours (e.g. 01:30 – 4:30 corresponds to the WS at the nominal time of 03:00), we have chosen to average the AOT data over the hour before and after each nominal WS hour. For example, the AOT value corresponding to the WS value at the nominal time of 03:00 is the average of the AOT from 02:00 – 03:00 and 03:00 – 04:00. Unless otherwise noted, AOT values throughout this study correspond only to times when WS data were available (i.e. daylight hours).

3. Results
3.1 Weather States
Figure 2 shows the distribution of the WSs monthly at each site for 2000-2012. FRW is the most common at both sites year-round, with frequencies within each month in the range of 19.9-42.5% at PSU and 32.1-47.2% at BER. The second most frequent WS at both sites was CIR,
which exhibited opposite behavior in its seasonality at the two sites. CIR shows a winter/spring maximum in frequency of occurrence at PSU (28.8% of cases for DJF and 25.9% for MAM), while the opposite behavior is observed at BER (i.e. a summer maximum with 43.6% of all WSs in JJA being CIR followed by 31.0% for SON). In contrast, SHC shows the reverse behavior at both sites, with a summer maximum at PSU (9.3% of all WSs in JJA) and winter/spring maximum at BER (9.5% for DJF and 9.9% for MAM).

One cloud type that has been well-studied for aerosol-cloud interactions is stratocumulus clouds (Wood, 2012). While not as abundant as other cloud types at the two sites, STC also show interesting seasonality. At Bermuda, STC is infrequent during the summer, accounting for only 0.1% of all observed WSs in August, but is much more common during the winter months, representing 10.7% of all observed WSs at Bermuda in December. However, at PSU the seasonal cycle is not as pronounced, with the prevalence of STC in the range of 1.3-4.8% of all WSs for each month throughout the year. Similarly, DCN, MDS, and MID show relatively little change seasonally at PSU. This behavior is observed at BER for DCN and MDS as well, but MID shows a much more pronounced seasonal cycle at BER, decreasing from a maximum of 11.2% in DJF to only 1.0% in JJA. Finally, CLR and PLR are much less frequent at BER as compared to PSU. The frequencies of occurrence for each WS by site and month can be found in Table S1 in the supplement. Although differences in frequency and seasonality were observed for the WSs at the two sites, the next section investigates the degree of consistency of the radiative environment of the WSs across the two locations.

3.2 Weather States and Radiative Properties

Previous studies have indicated that different WSs/cloud regimes exhibit unique radiative properties using radiative data from satellite-borne instruments (Oreopoulos et al., 2016;
Nevertheless, a key objective of this study is to connect the satellite-derived regimes to ground-based measurements. In order to investigate whether ground-based measurements identify unique radiative environments amongst the various WSs, two different derived quantities were examined.

Figures 3-5 show joint histograms of fractional sky cover and effective SW cloud transmissivity (SWdn/clear-sky SWdn) for each WS. As the joint histograms show similarities in the range of the values observed for both quantities at each site within each WS, these histograms lend insights into the properties of the clouds observed from the ground within each WS and their impact on the surface irradiance. CLR exhibits low values of fractional sky cover (average of 0.09 at PSU and 0.14 at BER) and high values of effective SW cloud transmissivity (average of 0.97 at PSU and 0.96 at BER), demonstrating that the satellite-retrievals are representative of the behavior in the ground-based measurements at both sites. The opposite behavior is observed for MDS, DCN, and MID at both sites, with the joint histograms showing high values of fractional sky cover (averages for these three WSs ranging from 0.94 – 0.96 at PSU and 0.92 – 0.98 at BER) and lower values of effective SW cloud transmissivity (averages of 0.33 – 0.41 at PSU and 0.35 – 0.48 at BER). FRW and CIR show a broad range of values for both ground-based quantities, although higher values of effective SW cloud transmissivity (averages for FRW/CIR of 0.74/0.77 at PSU and 0.83/0.79 at BER) do tend to be more common. In agreement with the ground-based data, FRW properties from the ISCCP data exhibit a very broad range of both cloud optical depths and cloud heights. While SHC also covers a broad range of values for both quantities, higher values of fractional sky cover (average of 0.75 at PSU and 0.74 at BER) are observed. Similar behavior is seen for STC, which shows even greater tendency to have higher values of fractional sky cover (averages of 0.87 at both sites). Finally, the PLR WS also covered the full range of fractional sky coverage and effective SW cloud transmissivity, but mainly at PSU owing to such sparse incidents of this WS observed over BER, with only 14 occurrences of PLR at BER in the 13-year period of study.
Figure 3. Joint histograms of effective SW cloud transmissivity and fractional sky cover for DCN, MDS, and CIR. The percentage listed next to the WS in each panel title represents the overall frequency of occurrence for that particular WS at each site for 2000-2012. The color scale in each WS figure represents the relative frequency of occurrence (RFO) for that particular set of conditions within that specific WS.

Figure 4. Same as Figure 3 except for PLR, MID, and FRW.

Figure 5. Same as Figure 3 except for SHC, STC, and CLR.
As demonstrated in Figures 3-5, the WSs appear to capture the distinct differences in the derived quantities from the ground-based radiation measurements that correspond well with the cloud properties from the satellite measurements. Furthermore, these differences appear to be consistent at the two sites within the domain given that the joint histograms at both sites appear similar for each WS.

### 3.3 Weather States and Aerosol Properties

#### 3.3.1 Average Aerosol Characteristics

In addition to describing cloud properties as measured from the surface for the various WSs, another goal of this study is to better understand whether the varying meteorology in these regimes correlates to distinct differences in aerosol characteristics of the regimes, of relevance for aerosol-cloud interactions. Figure 6 shows the seasonal averages and standard deviations of AOT from MERRA-2 at the two sites for each individual WS. The overall average AOT for each season is indicated with a dashed black line, and shows a strong seasonality at PSU, peaking in JJA (average AOT = 0.27) with a minimum in DJF (average AOT = 0.13). In contrast, BER shows a more consistent profile throughout the year, with a smaller peak in MAM (average AOT = 0.16) and minimum in SON (average AOT = 0.11). These trends agree with Figure 1 and previous seasonal analysis of MERRA-2 AOD showing higher values over the eastern coast of the U.S. in summer and higher values near Bermuda in spring (Figure 5 of Sorooshian et al., 2020). Furthermore, SO$_4$ is the dominant AOT component at PSU, while SS is much more influential at BER (Figure 6). Given the locations of these two sites (i.e. PSU near anthropogenic sources of the eastern United States and BER over the open ocean), the dominant AOD components for each site match expectations.

Interestingly, the various WSs seem to exhibit differences in their AOT levels throughout the course of the year. For example, at both sites, CLR tends to have the lowest values of AOT throughout the course of the year, with averages below that of each seasonal average. At PSU, the average AOT during CLR ranges from 0.06 (DJF) – 0.16 (JJA), while at BER the average AOT during CLR ranges from 0.07 (DJF) – 0.13 (MAM). AOT is strongly dependent on RH due in part to the hygroscopic growth of humidified particles. Furthermore, past work has shown positive correlations between RH and secondary species such as sulfate and nitrate (e.g. Tai et al., 2010). While not a direct comparison because AOT is a columnar measurement, ground-based measurements of RH co-located with the radiation measurement sites are available. As shown in Figure 6, CLR has seasonal average RH values (41.57 – 64.24% at PSU; 62.88 – 69.80% at BER) below those of the corresponding seasonal averages inclusive of all WSs (58.73 – 66.60% at PSU; 70.19 – 75.30% at BER) at both sites for all seasons. This difference is especially pronounced at PSU in DJF, MAM, and SON. In contrast, both sites exhibited year-round above average values of seasonal RH for MDS (74.59 – 79.79% at PSU; 78.52 – 83.13% at BER) and DCN (72.44 – 77.43% at PSU; 81.93 – 85.12% at BER). Both MDS and DCN also exhibit AOT values higher than the seasonal average at both sites year-round (Figure 6).

As stated in Section 2, we have chosen to use a reanalysis product for the AOT since remotely sensed retrievals of AOT are not possible during periods with high cloud fraction. Even during times with lower cloud fractions, issues related to cloud-contamination can exist, although some studies have successfully corrected for this effect (Ten Hoeve and Augustine 2016). Given the results of Figure 6, which shows a distinct difference between CLR and other WSs, our choice to use reanalysis products as opposed to remotely sensed data (which would only be
available during CLR periods) seems warranted. These results additionally challenge the frequently made assumption in aerosol-cloud-interaction studies using remote sensing that the aerosol observed in clear-sky pixels represents the aerosol environment in adjacent cloudy pixels. The sensitivity of aerosol-cloud relationships to various factors, including meteorological forcing and the history of air parcels (Loeb and Manalo-Smith, 2005; Mauger and Norris, 2007), complicates reaching robust conclusions in the current dataset about causality, and thus it is important to emphasize that this work is focused on relationships that can be investigated in more detail in future studies.

**Figure 6.** Seasonal averages and standard deviations of MERRA-2 AOT and measured surface level relative humidity (RH) for each WS. The dashed black lines represent the seasonal averages for all WSs at each site for RH and total AOT. The species comprising total AOT are black carbon (BC), dust, organic carbon (OC), sea salt (SS), and sulfate (SO₄). No evidence for AOT is shown for BER; only 14 instances of this WS occurred over the 13-year period examined, leading to seasonal average fractions for the species that are highly variable. Evidence of this variability can be seen in the very large contribution of dust to the average PLR AOT at BER in JJA (Figure 6), which was based on only two occurrences of PLR at BER in JJA. Both BC and dust show trends in fractional contributions that appear consistent across both sites. Previous studies have shown reductions in trade cumulus (Ackerman et al., 2000) in regions where absorbing aerosols are present. At both BER and PSU, the average percent contributions of BC and dust to the total AOT were consistently greater during CLR than the seasonal average contributions. Although the magnitude of contribution varied between the two sites, at both sites the greatest percentage point differences between the seasonal average and CLR average occurred in MAM for dust and SON for BC (Figure 7). While specific cases are not examined in
this study, it is plausible that the increased BC concentrations found at both sites during CLR, as opposed to other WSs, could indicate the influence of the aerosol semi-direct effect (Hansen et al., 1997) on clouds. However, the net impacts (i.e. increases or decreases in cloudiness) of the semi-direct aerosol effect have been shown to be strongly dependent on the vertical distribution of absorbing aerosols and cloud type (Koch and Del Genio, 2010). In addition, at both sites the fractional contribution of dust AOT is slightly above the seasonal average during the CIR for all seasons except JJA at BER. Dust has been shown in many previous works to be a good source of ice nuclei for cirrus clouds (e.g. Cziczo et al., 2013; DeMott et al., 2003). Other work has shown differences in correlations between dust and cloud amount depending on both the cloud type and geographical location (Mahowald and Kiehl, 2003).

![Figure 7](image)

**Figure 7.** Average and standard deviation of the fractional contribution of each component species to the total AOT for the various WSs at both PSU and BER in each season. The component species are black carbon (BC), dust, organic carbon (OC), sea salt (SS), and sulfate (SO$_4$). The dashed black lines represent the seasonal average contributions of each species to the total AOT for both sites. Note that PLR at BER is not shown due to the low number of instances of occurrence (14 total over a 13 year period) that led to high variability in the fractional contributions.

In contrast to the results for BC and dust, the profiles shown for OC and SS show less consistent seasonal patterns among the WSs at each site. This appears to be the case for SO$_4$ at BER as well, but a much stronger trend is present for SO$_4$ at PSU. In-cloud aqueous processing can contribute substantially to atmospheric production of SO$_4$ (Barth et al., 2000; Ervens, 2015), which may partially explain the higher contributions of SO$_4$ to the total AOT during cloudier WSs (i.e. MDS, MID, SHC, STC) at PSU, especially as compared to CLR at PSU. Furthermore, SO$_4$ can act as effective cloud condensation nuclei (CCN) although previous studies over the northeastern Atlantic seaboard of the U.S. (Hegg et al., 1995) showed that SO$_4$ cannot account for the full CCN budget. This may explain inconsistent patterns observed at BER in SO$_4$ (which has much lower contributions to total AOT) as compared to PSU, where certain cloud types
presumably exhibit higher influences from \( \text{SO}_4 \). However, for each of these relationships analyzed between the various aerosol types and the WSs, additional factors such as meteorological covariation and the vertical distribution of aerosol may also influence the observed relationships.

### 3.3.2 Extreme Aerosol Events

In addition to average aerosol characteristics, examining the prevalence of high and low aerosol events within each WS can lend additional insights. Similar to the methodology of Gryspeerdt et al. (2014), high and low aerosol events were isolated in order to examine the relative frequencies of different cloud regimes for each event type. The threshold for high (low) AOT events was set separately at PSU and BER as the 90\textsuperscript{th} (10\textsuperscript{th}) percentile of all AOT values (Table 2) within each season corresponding to instances with available WSs (i.e. daylight hours).

**Table 2.** Thresholds for high (90\textsuperscript{th} percentile of MERRA-2 AOT) and low (10\textsuperscript{th} percentile of MERRA-2 AOT) aerosol events within each season at PSU and BER.

<table>
<thead>
<tr>
<th>Season</th>
<th>PSU 90th Percentile</th>
<th>PSU 10th Percentile</th>
<th>BER 90th Percentile</th>
<th>BER 10th Percentile</th>
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</thead>
<tbody>
<tr>
<td>DJF</td>
<td>0.234</td>
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</tbody>
</table>

Figure 8 shows the relative frequency of occurrence (RFO) of the AOT corresponding to each WS in every season falling above or below the high or low aerosol event threshold, respectively. Values above the zero line indicate the RFO for high AOT events (RFO\textsubscript{high AOT}), while values below the zero line indicate the RFO for low AOT events (RFO\textsubscript{low AOT}). For example, of the 446 instances when MDS occurred at PSU in DJF, 99 of those instances had AOT values greater than the 90\textsuperscript{th} percentile of all AOT values at PSU in DJF, resulting in an RFO\textsubscript{high AOT} of 0.222 that is displayed above the zero line. In contrast, only 7 instances of MDS at PSU in DJF fell below the low aerosol event threshold; the corresponding RFO\textsubscript{low AOT} of 0.016 is displayed below the zero line. In addition, PLR occurred only once at BER in SON; this instance also fell into the low aerosol event category, resulting in an RFO\textsubscript{low AOT} = 1.
Relative frequency of occurrence (RFO) for instances where the AOT values corresponding to each WS fall within the top 10% (high AOT) or bottom 10% (low AOT) of AOT values at each site within each season. Note that all values of RFO were less than or equal to 0.5 except for PLR in SON at BER, which had an \( \text{RFO}_{\text{low AOT}} = 1 \) based on one occurrence of this combination of factors. This discrepancy is indicated with an arrow on the respective bar, since the axis extends only to 0.5 in order to increase figure readability for the other bars.

As shown in Figure 8, CLR consistently demonstrates the highest RFO for low aerosol events at PSU (0.28 – 0.45) and BER (0.19 – 0.33), with the singular exception being the special circumstances of PLR at BER in SON. The higher incidence of low aerosol events within CLR is at least consistent with a lack of in-cloud aqueous secondary aerosol formation. However, the influence of wet-scavenging is less clear since the presence of cloud-containing WSs would be required for the process to occur. While precipitation would not be present during CLR periods, wet-scavenging could have previously occurred during other WSs, leading to both a depletion in aerosol and a loss of cloud cover. Furthermore, the lower values of RH found during CLR could also drive the values of AOT lower than would be the case during cloudy periods (Figure 6). In addition, CLR exhibited very low values of \( \text{RFO}_{\text{high AOT}} \), with a maximum of 0.02 at PSU in JJA and 0.03 at BER in MAM. FRW and CIR also each yielded more events in the low AOT category than high AOT for each season except MAM at PSU for FRW and SON at BER for CIR.

In contrast, MDS, DCN, MID, and SHC all demonstrate higher \( \text{RFO}_{\text{high AOT}} \) than \( \text{RFO}_{\text{low AOT}} \) at both sites year-round. Excepting the unique circumstance of PLR at BER, the highest \( \text{RFO}_{\text{high AOT}} \) occurred for SHC at BER in JJA, with 42 of the 94 instances of SHC at BER during JJA being in the high AOT category. This is interesting given that SHC occurs infrequently at BER during the summer months (Figure 2). STC also exhibits behavior typically yielding a higher RFO for high AOT events than low AOT events, with the two exceptions being during DJF at BER and during JJA at PSU when there were an equivalent number of high and low AOT events for STC. These combined results broadly point to the complexities of aerosol-cloud-interactions and the ways in which different cloud types can impact aerosol and also how aerosol
impact clouds. For example, Koren et al. (2005, 2010) showed that clouds developing in high AOD environments were more convective, leading to greater cloudiness and lower cloud top pressure. In this case, some of the WSs with higher cloud fractions, such as MDS, DCN, and MID (Figures 3-4), demonstrate consistently higher $RFO_{\text{high AOT}}$ than $RFO_{\text{low AOT}}$. The time evolution of various WSs within the different aerosol environments is discussed subsequently.

Building on the methodology of Gryspeerdt et al. (2014), Figure 9 shows the 3 hr transitions between WSs, separated into various AOT categories. In addition, Figure S2 in the supplement shows the difference in transitions between the high and low AOT categories in Figure 9. The AOT categories were determined based on conditions for the starting WS using the seasonal limits in Table 2. As was the case for the regimes derived via MODIS data in Gryspeerdt et al. (2014), most WSs remained the same such that no transition to a different WS occurred. However, there are some notable exceptions to this general rule. Under low AOT conditions, FRW was much more likely to transition to CLR than under high AOT conditions. While this behavior is much more pronounced at PSU, a similar trend can be seen at BER. Furthermore, with the single exception of the low AOT category at PSU, the most common transition from a starting WS of CLR was to FRW, not CLR. Consistent with the results in Figure 8, CLR very infrequently fell into the high AOT category; however, interestingly for any starting WS in the high AOT category, CLR was almost never the ending WS (only 1 instance at PSU and 4 at BER). Therefore, it appears that CLR was very unfavorable to both existence and formation under high aerosol conditions. In contrast, MDS, DCN, MID, and SHC showed opposing behavior, with especially favorable ending states under high aerosol conditions and low occurrence under low AOT conditions. As was also shown in Figure 8, the prevalence and evolution of more convective cloud types under high AOT conditions shown here agree with previous studies from the region (Koren et al., 2005, 2010). Previous studies have also indicated a link between higher AOT and higher cloud fractions (e.g. Loeb and Schuster, 2008) and Quaas et al. (2010) outlined six potential hypotheses for these correlations, including aerosol size changes from increased humidity near clouds. Under low AOT conditions, the most favorable ending states at the two sites were FRW and CIR. In addition, CLR was a prevalent starting and ending state under low AOT conditions at PSU, but not at BER. Transitions associated with SHC and STC WSs are important with regard to ongoing research focused on analyzing the evolution of these cloud types. Previous work in the North Atlantic has shown that models underrepresented the prevalence of the shallow cumulus WS (Rémillard and Tselioudis, 2015). Extensive past work has probed factors leading to STC transitioning to SHC (e.g. Yamaguchi et al., 2017). For the cases analyzed in this study, SHC tends to be a more favored ending state under high aerosol conditions as opposed to low aerosol conditions.

Given the various feedbacks in aerosol-cloud interactions, it is difficult using this dataset to identify the exact mechanisms leading to the observed relationships between aerosols and cloud types in the region. However, these results indicate that the various WSs can provide an effective background for studying aerosol-cloud interactions given the differences observed among the WSs under varying aerosol conditions at the two sites and can guide analysis of more detailed data sets from airborne campaigns in the region.
Figure 9. Frequency of transitions between WSs for 3 hr intervals. The low, middle, and high AOT categories are determined seasonally using the 10th and 90th percentile limits at each site in Table 2, with total counts compiled across all seasons for each category. The AOT categories are determined based on the AOT for the starting WS. The color scale represents the relative frequency of occurrence for the transition from a starting WS to the subsequent ending WSs 3 hr later. Note that the frequency sums to a total of one for each row (excluding the first column for the total instances of the starting WS). The total number of times each combination of events and conditions are present is also shown.

4. Conclusions

The atmospheric properties of various WSs derived from satellite retrievals were investigated using remotely sensed ground-based data and reanalysis results at two sites in the WNAO region: PSU and BER. At both sites, FRW was most common, followed by CIR. The CIR WS exhibited different seasonality at the two sites, with a higher prevalence in the winter months at PSU as opposed to a peak during the summer months at BER. However, at both sites, the ground-based retrievals of fractional sky cover and effective shortwave cloud transmissivity showed similarity between the two sites for each WS. Furthermore, between the various WSs, clear differences in the distributions of these retrievals were observed. For example, the CLR WS was effectively captured in the ground-based retrievals through low observed values of fractional sky cover and high observed values of effective shortwave cloud transmissivity at the two sites.

With the connections between the satellite-derived WSs and ground-based radiative properties established, the aerosol properties of the various WSs were examined. Due to the locations of the two sites of interest (PSU on the North American east coast and BER over the remote ocean), the typical influencing sources of atmospheric aerosols at the sites were quite different. However, even with these differences in location, similarities in the aerosol properties among the various WSs were clearly observed. The clear sky WS exhibited below-average AOT values for each season at both sites; furthermore, this WS exhibited consistently high coincidence with periods of AOT falling within the bottom 10% of the seasonal values at each site. In contrast, other WSs such as MDS and DCN showed above-average AOT values for each
season at both sites. In addition to total AOT, the changes in speciated AOT were examined for the various WSs. At both sites, CLR had above-average contributions from BC and dust to the total AOT year-round. OC and SS showed few differences in contributions to the various WSs. While this was also true of SO\textsubscript{4} at BER, PSU showed distinct trends in the contribution of SO\textsubscript{4} to the various WSs. In particular, MDS, MID, SHC, and STC had higher fractions of SO\textsubscript{4} year-round as compared to the seasonal averages.

Finally, the time evolution of various WSs was examined in the context of high and low aerosol events at the two sites. At both sites, the general trend was for the WS to remain the same after the 3 hr interval; however, notable exceptions to this trend existed. CLR frequently evolved to FRW as opposed to remaining CLR. Furthermore, CLR was neither a very frequent starting nor ending state under high AOT conditions. In contrast, MDS, DCN, MID, and SHC were more favored as ending states under high AOT conditions.

Overall, the results of this study indicate the various atmospheric conditions present under the satellite-derived WSs. These WSs have previously proven useful for evaluating the performance of various models in reproducing cloud schemes; these results indicate the WSs could potentially be used to constrain the study of aerosol conditions as well. Further studies are needed to identify other regions where differences in aerosol conditions are observed amongst the various WSs and whether ground-based aerosol measurements, as opposed to the modelled AOT used in this study, also exhibit differences amongst the various WSs. In addition, studies further analyzing aerosol characteristics during WS transitions, especially the aerosol speciation in a variety of locations worldwide, could lend additional insights to the relationships between aerosol and the WSs.

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**Data Availability:** WS data were obtained from the NASA Goddard Institute for Space Studies ISCCP website (https://isccp.giss.nasa.gov/wstates/hggws.html). Relative humidity and radiation data from PSU and BER, including the derived quantities used in this study, and AOD data from PSU were downloaded from the NOAA Earth System Research Laboratories Global Monitoring Laboratory SURFRAD website (https://www.esrl.noaa.gov/gmd/grad/surfrad/index.html). AERONET data from the sites on Bermuda were obtained from the NASA AERONET site (https://aeronet.gsfc.nasa.gov/). MERRA-2 AOT data were obtained from the NASA Goddard Earth Sciences Data and Information Services Center (https://disc.gsfc.nasa.gov/).

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