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Atmospheric Research Over the Western North Atlantic Ocean Region and North American East Coast: A Review of Past Work and Challenges Ahead

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Abstract

Decades of atmospheric research have focused on the Western North Atlantic Ocean (WNAO) region because of its unique location that offers accessibility for airborne and ship measurements, gradients in important atmospheric parameters, and a range of meteorological regimes leading to diverse conditions that are poorly understood. This work reviews these scientific investigations for the WNAO region, including the East Coast of North America and the island of Bermuda. Over 50 field campaigns and long-term monitoring programs, in addition to 715 peer-reviewed publications between 1946 and 2019 have provided a firm foundation of knowledge for these areas. Of particular importance in this region has been extensive work at the island of Bermuda that is host to important time series records of oceanic and atmospheric variables. Our review categorizes WNAO atmospheric research into eight major categories, with some studies fitting into multiple categories (relative %): Aerosols (25%), Gases (24%), Development/Validation of Techniques,

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Models, and Retrievals (18%), Meteorology and Transport (9%), Air-Sea Interactions (8%), Clouds/Storms (8%), Atmospheric Deposition (7%), and Aerosol-Cloud Interactions (2%). Recommendations for future research are provided in the categories highlighted above.

1. Introduction

Decades of atmospheric research have targeted the Western North Atlantic Ocean (WNAO) region, which we define here as including the oceanic domain bounded by 25–50°N and 60–85°W, and North America's East Coast (Figure 1). Shipboard, airborne, ground, and satellite measurements in addition to modeling studies have been used to characterize the WNAO's atmospheric profile, including gas/aerosol/precipitation chemistry, cloud and storm features, boundary layer structure, and transport patterns. This diverse ocean basin presents a complex atmospheric system with many unknowns, even with the substantial investment of field measurement campaigns and programs focused on the region over the last five decades.

A disproportionately large amount of research has focused on the WNAO in contrast to most other oceanic basins largely owing to its accessibility and being downwind of the polluted East Coast of North America (Fehsenfeld et al., 2006), which provides several advantages for atmospheric research due to the importance of gradients in studying different phenomena. The majority of the WNAO research has focused on atmospheric chemistry issues, as transport and aging can be studied effectively with anthropogenic plumes advected through a cleaner marine backdrop where sources of pollution are much more limited (Fehsenfeld et al., 1996). The broad North American urban plume is sufficiently strong that it has an appreciable influence on western Europe through surface ozone (O₃) concentrations (Li et al., 2002) and processes such as wet deposition (Tarrason & Iversen, 1992). In addition to pollution gradients being an important feature of the WNAO region, the reduction in pollution since the 1980s (Feng et al., 2019; Frost et al., 2006; Jongeward et al., 2016; Yoon et al., 2014; Zhang & Reid, 2010; Zhao et al., 2008) owing to regulatory activities (Smith et al., 2001; Streets et al., 2006) now affords a prime opportunity to contrast current research of pollution-dependent topics with documented results from past decades to gain new insights.

It is convenient timing to review what is known about the WNAO region and North American East Coast to provide context for ongoing and future research activities. In response to this need, this paper consolidates and synthesizes past research relevant to the WNAO region, including the North American East Coast and Bermuda, and highlights pressing knowledge gaps that require attention. This review covers a wide range of atmospheric topics, including gases, aerosols, wet deposition, cloud and storm features, aerosol-cloud interactions (ACI), air-sea interactions, meteorology and transport studies, and also works that leveraged special features of the WNAO to develop and/or validate new techniques, instruments, models, and retrievals. The order of sections in this review is as follows: (i) meteorological features and atmospheric circulation over the WNAO, (ii) a chronological history of campaigns and other measurement or modeling efforts, (iii) a synopsis of decades of research conducted at Bermuda and the adjacent Sargasso Sea, and

(iv) a synthesis of previous results and discussion of remaining scientific questions and challenges for continued research in the study region.

2. Atmospheric Circulation and Transport

The primary atmospheric phenomena investigated in past research for the WNAO region and East Coast (e.g., atmospheric chemistry, boundary layer structure, clouds and precipitation, air-sea interactions) depend on meteorology. The transport and spatial distribution of gases and aerosols are especially sensitive to the meteorological setting at any moment in time. To put the subsequent discussion of past research in proper context, an overview is provided for the dominant regional-scale atmospheric features in the study region. The reader is referred to several works for snapshots of specific meteorological and transport conditions for individual field campaigns to be described later (e.g., Angevine et al., 1996; Fuelberg et al., 2000; Fuelberg et al., 2007; Merrill & Moody, 1996; Moody et al., 1996; Sethuraman et al., 1986; Stunder et al., 1987).

A central climatological feature of the North Atlantic is the presence of a semi-permanent high pressure, commonly referred to as the Azores or Bermuda High (e.g., Davis et al., 1997) (Figure 2, black contours). The Bermuda-Azores High's maximum development occurs in summer, with a pattern that drives a well-defined anticyclonic circulation in the boundary layer, with easterly trade winds south of 25°N that progressively move northward as they reach the continent, and southwesterlies nearly parallel to the coastline north of 25°N (Figure 2, June-July-August (JJA), green arrows and colored contours). In contrast, the strengthening of the subpolar low pressure (Icelandic Low) in the extra-tropics, starting in autumn, limits the expansion of the Bermuda-Azores High, while inducing westerly winds in the boundary layer, with a northerly component near the continent in winter, and trade winds detached from the circulation pattern farther north. In addition, the low and mid-tropospheric circulation is characterized by westerly winds (Figure 2, winds at 700 hPa, gray arrows), particularly during winter, when midlatitude weather disturbances are a dominant feature over the region.

Concomitant changes in the Bermuda-Azores High and Icelandic Low, in connection with the North Atlantic Oscillation (NAO), are a central aspect of the North Atlantic climate, as well as key in determining transport pathways between North America and Europe (Christoudias et al., 2012; Creilson et al., 2003; Li et al., 2002). The concept of NAO refers to a "large-scale alternation of atmospheric mass between the North Atlantic regions of subtropical high surface pressure (centered near the Azores) and subpolar-low surface pressure (extending south and east of Greenland)" (Lamb & Pepler, 1987). The NAO involves the meridional atmospheric pressure difference between the subtropical and subpolar Atlantic Ocean, with the NAO index typically defined as the sea level pressure difference between Lisbon (Portugal) or Ponta Delgada (Azores) and Stykkisholmur/Reykjavik (Iceland) (Hurrell, 1995). The positive NAO phase is characterized by a higher than average pressure difference (i.e., strong Icelandic Low and Bermuda-Azores High) and anomalous strong westerlies across the North Atlantic. In contrast, the negative NAO phase is associated with weakened Icelandic Low and Bermuda-Azores High, resulting in weaker westerlies. The NAO is especially prominent during winters, but is present throughout the

year. Decadal variability in the NAO has become more pronounced since 1950 according to Hurrell (1995); this trend has implications for intercontinental pollution transport across the North Atlantic Ocean between Europe, North America, and the Arctic.

The WNAO region sees its share of tropical cyclones (TC) (Landsea & Franklin, 2013), with approximately 101 storms that either tracked through the region or made landfall on the United States (U.S.) East Coast from 1950–2017. Colbert and Soden (2012) looked at various climate features that can affect TC tracks over WNAO. One of these is the Bermuda-Azores High, whose strength and location influence large-scale steering flow. Other factors that can influence TC formation and tracks include: El Niño Southern Oscillation (ENSO), NAO, and phase of the Atlantic Meridional Mode. The National Hurricane Center's North Atlantic hurricane database (HURDAT) (Landsea & Franklin, 2013) classifies TC tracks into one of three categories. Two of these are of interest for the WNAO region: (i) recurving landfall TCs that threaten the U.S. East Coast, and (ii) recurving ocean TCs that recurve into the open ocean but could still influence the atmospheric conditions in the WNAO region, including Bermuda (Fig. 1 in Colbert & Soden, 2012). Nakamura et al. (2009) classified North Atlantic TC tracks using a clustering method, which provides a way to assess spatial characteristics (i.e., track shape, genesis location, seasonality, and landfall). There are six clusters, three of which included TCs that originated, tracked through, or made landfall in the WNAO region. Cluster 2 is of most importance since it included U.S. East Coast storms and also had the longest TC season (April to November).

Winter extratropical cyclones (ETCs) also pose a hazard to the U.S. East Coast (Hall & Booth, 2017). ETC winds can on occasion reach hurricane force and cause damaging coastal storm surges. Orton et al. (2016) found that in the New York City area, ETCs are a dominant cause of surge events that have an annual probability greater than 1%. The National Oceanic and Atmospheric Administration (NOAA) National Climatic Data Center (NCDC) estimated that from 1980 to 2016, major ETC events caused more than \$1 billion in damage for each event (<https://www.ncdc.noaa.gov/billions/summary-stats>). The importance of ETCs has led to a series of studies evaluating the effect of ENSO and NAO indices on heightened ETC activity (DeGaetano et al., 2002), increases in likelihood of storm surge (Bernhardt and DeGaetano 2012), and the probability of extreme ETC events (Hall and Booth 2016).

The aforementioned atmospheric and meteorological phenomena largely define the main gas and aerosol transport pathways over the WNAO. It is during the positive NAO phase that more North American pollution is found over the WNAO extending towards Europe, while during the negative NAO phase weaker westerlies disrupt the ability of North American pollution to flow eastward towards Europe. In addition to changes in the circulation patterns, the NAO has a discernible impact on temperature, precipitation, storm frequency, and budgets of aerosols and gases such as O₃ and carbon monoxide (CO) (e.g., Appenzeller et al., 2000; Eckhardt et al., 2003; Hurrell, 1995; Li et al., 2002; Ma et al., 2013). In addition to the NAO, the Bermuda-Azores High, the Icelandic Low, and the trade winds in the subtropics all impact transport pathways. For instance, the anticyclonic circulation in summer promotes transport of aerosols from the Eastern Atlantic and North Africa to the WNAO (Chen & Duce, 1983; Jickells et al., 1998). In winter and spring, the prevalent tropospheric winds favor pollution transport from North America over the Atlantic Ocean

and towards Europe. Winter-spring westerly flow from the East Coast is assisted by some combination of three synoptic mechanisms according to Creilson et al. (2003): (i) westerly flow of boundary layer air, (ii) westerly transport of free tropospheric air that was convectively lofted above land, and (iii) frontal lifting owing to the warm conveyor belt process. Midlatitude cyclones are critical in providing sufficient energy for pollutants from North America to travel eastward to the WNAO (Cooper et al., 2002a; Cooper et al., 2002b; Cooper et al., 2001), with the classic midlatitude cyclone being comprised of the following airstreams (Carlson, 1998): warm conveyor belt (WCB), cold conveyor belt (CCB), dry airstream (DA), and post cold front (PCF) airstream. Once air is over the WNAO, large-scale features over the WNAO take control that govern the subsequent path of the air.

An interesting case study is highlighted here that shows the complexity of weather and climate patterns at just one point location along the North American East Coast. Keim et al. (2005) categorized synoptic weather patterns ('weather types') over New England into nine types and used them to find relationships between weather conditions and $PM_{2.5}$ (i.e., mass concentration of particles with aerodynamic diameters less than $2.5 \mu m$) at a rural site in New Hampshire. Many patterns existed for the study region owing to features such as extremes of high and low temperatures, complex terrain, coastal positioning, latitude ($\sim 45^\circ N$), intense rainfall, blizzard conditions, droughts, and significant inter-diurnal changes in temperature (Zielinski & Keim, 2003). Their classification system relied on an atmospheric pressure-based system where a particular weather type would exist based on geographic proximity to centers of low and high pressure associated with cyclones and anticyclones, respectively, and frontal boundaries (Muller, 1977). The extremes of their identified weather types included the Canadian High (on eastern side of anticyclone – driest/coldest air in winter from north/northwest) and the Frontal Atlantic Return (in warm sector of cyclone - hot and humid air from south/southwest with clouds/rain). The other seven regimes exhibited a distinct air flow pattern associated with a specific temperature range and varying conditions of clouds and precipitation; three examples include the Gulf of Maine Return (on southern side of anticyclone), the New England High (coincident with center of high pressure), and the Atlantic Return (on western side of anticyclone). Their general conclusion from relating the nine weather patterns to $PM_{2.5}$ levels was that northerly and northwesterly flow coincided with low $PM_{2.5}$, while southerly and southwesterly flow yielded the highest $PM_{2.5}$ levels.

3. History of WNAO Research

The WNAO region represents an area with diverse conditions associated with atmospheric chemistry, meteorology, and clouds. For instance, gradients in chemistry are provided by the pollution emanating from North America and Europe, and dust from North Africa, North America, and Asia. Based on the literature, it is the transport and evolution of pollution over the WNAO that has garnered most interest of all research areas for the WNAO, for which a conceptual graphic is shown in Figure 1. As will be shown, most field studies have focused on the summer period. There was early recognition (e.g., Husar et al., 1981) of the mounting evidence of regional haziness over the Eastern U.S. peaking in that season, which would later be linked to factors such as more combustion emissions (e.g., biomass burning) and enhanced photochemistry (e.g., Hand et al., 2019).

For historical perspective of studies examining various aspects of atmospheric research, a timeline of major measurement efforts relevant to the WNAO is provided here, beginning when activities started to accelerate in the 1970s. Owing to the significance of North American pollution outflow to the WNAO region, studies are reviewed over the North American East Coast; studies based farther inland or with a focus over the Gulf of Mexico have been omitted for this review. The following five sub-sections examine each decade of research from the 1970s to the present, based on the date of when a particular measurement campaign or monitoring program was initiated rather than when the results were published, which often occurred multiple years later. Owing to extensive research conducted just for Bermuda and the surrounding Sargasso Sea (~20% of the 715 publications), those works are reviewed separately in Section 4.

Figure 3 provides a timeline of named field campaigns and Table 1 shows a breakdown of peer-reviewed publications into the following research themes: (i) gases, (ii) aerosols, (iii) wet deposition, (iv) cloud and storm properties, (v) ACI, (vi) air-sea interactions, (vii) any aspect of a new method, model, or instrument and/or validation studies between observations and models or retrievals, and (viii) meteorology and air flow analysis. The rationale for this categorization is based partly on the higher likelihood of success to accurately classify published works. An alternative categorization scheme for (i-iii) could potentially have been based on subsets of species (e.g., sulfur, nitrogen, organic matter, mineral aerosol, primary marine aerosol production) cycling between the three phases; however, more subjective reasoning and thus error in categorization would result for the individual works which measured a large suite of species but focused more on only one of the subsets of species. Table S1 shows an extended version of Table 1, listing each publication categorized into the same eight research themes. Some publications were cross-cutting and were assigned to multiple categories. More specifically, 298, 66, and 10 publications intersected two, three, and four different categories, respectively.

3.1 1970 – 1980

Beginning in the 1970s, efforts ramped up along the U.S. East Coast to monitor aerosols and gases. A multi-decadal time series of lidar data was initiated in 1974 at the National Aeronautics and Space Administration (NASA) Langley Research Center (LaRC) focused on stratospheric aerosols (Woods & Osborn, 2001). Highlights of that long-term dataset were evidence of disruptions such as major volcanic eruptions from Fuego (1974), El Chichon (1982), and Mt. Pinatubo (1991). Hoppel et al. (1984) characterized aerosol size distributions (0.006 – 2.2 μm radius) at Wallops Island, Virginia in June 1979 and contrasted results based on different air mass trajectories and whether there was marine or continental influence. They showed reductions in the number of small particles as the length of trajectories over water increased. Measurements in Miami, Florida as early as July – September 1974 confirmed the presence of Saharan dust transported across the Atlantic Ocean to North America (Savoie & Prospero, 1977); those early aerosol measurements were conducted as part of the Global Atmospheric Research Program Atlantic Tropical Experiment (GATE), which also included measurements at the Cape Verde Islands and Barbados, West Indies. Measurements at Nantucket Island, Massachusetts for June – August periods in 1975 – 1976 showed evidence of photochemical O_3 formation during transport

over the open ocean from the mid-Atlantic states based on biological indicators in the form of bioassays with O₃-sensitive tobacco plants (Kelleher & Feder, 1978).

There was growing interest in the issue of acid precipitation in the 1970s, with various investigators showing that in the Northeast U.S. (specifically New Hampshire) the pH of precipitation samples was consistently below the expected pH based on equilibrium with CO₂ (i.e., 5.6), with values frequently below 4.4 (Cogbill & Likens, 1974; Likens & Bormann, 1974; Likens et al., 1972). It was hypothesized at the time that the acids responsible were sulfur dioxide (SO₂) and nitrogen oxides (NO_x: NO and NO₂) (Galloway et al., 1976). Partly in response to these growing issues with acid rain, the Sulfate Regional Experiment (SURE) aimed to study the link between pollutants such as SO₂ and secondarily produced products such as sulfate (SO₄²⁻) over the northeastern U.S. (Perhac, 1978). Aircraft were used in intensive studies between 1977 and 1978, which helped to show that SO₂ and NO_x exhibited greater differences in their spatial distributions in contrast to secondarily formed aerosol products (Blumenthal et al., 1984). The airborne data also showed better mixing of pollutants during the day as compared to night, with particle number concentrations within the lower 1500 m more than twice as high during midday than at night or early morning hours, suggestive of significant photochemical production with daylight (Blumenthal et al., 1984).

The Western Atlantic Ocean Experiment (WATOX) began in 1979 to characterize the sources, evolution, and fate of air pollution outflow from North America (Galloway et al., 1988). Studies motivating WATOX showed that U.S. East Coast pollution outflow could be transported hundreds of kilometers to areas such as Nova Scotia (e.g., Beattie & Whelpdale, 1989; Brice et al., 1988; Kelleher & Feder, 1978; Spicer, 1982; Zeller et al., 1977). Trajectory analysis showed that ~60% of air masses arriving at Bermuda originated from direct flow off of North America (Miller & Harris, 1985). As wet scavenging is a major sink for many pollutants over the WNAO region, an extensive amount of studies have collected precipitation samples via ship measurements off the U.S. East Coast and at Bermuda (e.g., Church et al., 1982; Church et al., 1984; Jickells et al., 1982). WATOX had multiple intensive periods with airborne measurements conducted to further characterize the sources and vertical structure of gases and aerosols over the WNAO, including WATOX-85 (February – May 1985) (e.g., Galloway et al., 1988) and WATOX-86 (January – June 1986) (e.g., Galloway & Whelpdale, 1987). A selected result from these airborne efforts was that SO₂ concentrations decreased in the boundary layer by a factor of 20 from the East Coast to Bermuda, while SO₄²⁻ levels remained the same (Hastie et al., 1988). Additionally, boundary layer NO_x and nitric acid (HNO₃) levels decreased by an order of magnitude out towards Bermuda, while nitrate (NO₃⁻) dropped by a factor of two.

Overlapping in time and focus with WATOX was the Global Precipitation Chemistry Project (GPCP), which was initiated in 1979 and lasted until 1995. The period between 1980 and 1984 was of most relevance to the WNAO as precipitation samples were collected both onboard ships (Galloway et al., 1983) and at a surface site at St. Georges, Bermuda (Galloway et al., 1982). The consensus from many of these early studies characterizing wet deposition chemistry was that air masses originating over North America were coincident with high levels of SO₄²⁻, NO₃⁻, trace metals, and the hydrogen ion (H⁺) in rain, whereas

air masses from the Eastern Atlantic exhibited much lower background values (Galloway et al., 1983; Jickells et al., 1982).

3.2 1980 – 1990

The first airborne measurements of the vertical distribution of aerosols over the WNAO were executed in July 1981 and August 1982 between Wallops Island, Virginia, and Bermuda with a differential absorption lidar (DIAL) (Harriss et al., 1984). The data confirmed what was thought at that time from surface measurements, that North American pollution was being advected over the WNAO and impacting remote areas such as Bermuda. The momentum around the issues of North American outflow was growing rapidly at this time, as this phenomenon had already been linked to acid rain/snow, trace metal deposition, O₃ chemistry, and visibility in remote oceanic regions (e.g., Rahn & McCaffrey, 1979).

While there was already growing impetus in atmospheric chemistry research over the WNAO, the Mesoscale Air-Sea Exchange (MASEX) experiment in January 1983 was one of the first to gain insight into boundary layer convection and clouds in the marine boundary layer (MBL) (Melfi et al., 1985). This experiment's main objective was to examine air-sea interactions during Arctic cold air outbreaks (CAOs) (Boers & Melfi, 1987). MASEX provided some early demonstrations of the value of airborne lidar data in studying MBL structure, entrainment rate, wind shear, and relative heat fluxes (Melfi et al., 1985). Furthermore, various studies demonstrated the success of numerical models in reproducing the horizontal character of cloud fields, heat flux profiles, and mean temperatures during MASEX CAO events (Bechtold et al., 1992; Raasch, 1990). Closely related to MASEX was a separate research cruise in November 1983 off the North Carolina coast to study the thermal structure of the MBL (Sethuraman et al., 1986); those results showed that MBL was deeper with enhanced sensible heat fluxes during a CAO in contrast to other synoptic regimes encountered.

A multi-platform approach (ships, moorings, aircraft, satellites) was used during the Frontal Air-Sea Interaction Experiment (FASINEX) campaign between 1984–1986 to examine air-sea interactions in the subtropical convergence zone (STCZ) southwest of Bermuda (Charnock & Businger, 1991; Stage & Weller, 1986; Weller, 1991; Weller et al., 1995). FASINEX was designed to be over the open ocean to study the MBL structure's response to sea surface temperature (SST) gradients without influence from the transition from land to sea (Weller, 1991). FASINEX was conducted just prior to the Genesis of Atlantic Lows Experiment (GALE) in February 1986, which involved airborne flights to understand the nature of CAOs off the U.S. East Coast (Dirks et al., 1988). A number of GALE studies reported on results related to the nature of CAOs based on some combination of airborne, surface, and remote sensing data (Bane & Osgood, 1989; Vukovich et al., 1991). GALE was coordinated with the Canadian Atlantic Storms Program (CASP) field experiment, which was held during January – March 1986 to improve understanding of East Coast storm structure and dynamics (Stewart, 1991). Selected results from CASP included insight into how coastline structure governs storm features and that the nature of precipitation varied greatly (e.g., snow, freezing rain, ice pellets, rain) within each storm.

Similar in scope to WATOX studies, Church et al. (1991) reported on precipitation chemistry results (in addition to gas and aerosol) from a North Atlantic cruise from Senegal to Woods Hole, Massachusetts (April – May 1984). They concluded that air masses not contacting land for five days had a composition resembling that from a pristine marine atmosphere, with most of the non-sea salt (nss) SO_4^{2-} derived from oxidation of biogenic dimethylsulfide (DMS) (Church et al., 1991). Another set of cruises between 1985 – 1986 extending from the U.S. East Coast to the Sargasso Sea provided extensive results associated with gas, aerosol, and rain water properties over the WNAO (Berresheim et al., 1991). They reported on DMS fluxes from the open ocean as well as relative influences of different sources to atmospheric sulfur (S) and nitrogen (N) species.

The Global Change Expedition/Coordinated Air-Sea Experiment/Western Atlantic Ocean Experiment (GCE/CASE/WATOX) joint effort took place in July – September 1988 to understand biogeochemical cycling of major inorganic constituents (e.g., S, N), carbon (C), trace metals, and oxidants such as O_3 over the North Atlantic Ocean (Kim et al., 1990; Pszenny et al., 1990). The CASE component involved WATOX investigators focusing on a period of predominantly easterly flow impacting the WNAO, with CASE/WATOX being a part of the larger GCE organized by NOAA. A 49-day ship cruise was conducted from Norfolk, Virginia to Bermuda, Iceland, the Azores, and Barbados (GCE). Additionally, another cruise was conducted near Bermuda (WATOX), and flights were conducted off the Virginia Capes and near Bermuda (CASE/WATOX). By means of field measurements (Sievering et al., 1991) and modeling (Luria & Sievering, 1991), GCE/CASE/WATOX research showed that appreciable levels of SO_4^{2-} in WNAO sea salt particles originated from the heterogeneous conversion of SO_2 , which is greatly facilitated by the abundance of moisture and thus aerosol water (Kim et al., 1990). Size distribution measurements between 0.1 – 32 μm over the WNAO at altitudes of 150 and 2750 m could be fitted well with three lognormal distributions (Kim et al., 1990): “fine”, “large”, and “giant” modes with volume geometric median diameters of ~0.23, 1.42, and 5.46 μm , respectively. When those data were compared near off the U.S. East Coast versus the open-sea Bermuda area, fine mode volume concentrations were higher by the East Coast owing to enhanced anthropogenic pollution, while the giant mode was also enhanced by the East Coast due to enhanced relative humidity and wind speeds. The large mode showed the least difference between the two areas.

In the winters of 1988 and 1989, the Experiment on Rapidly Intensifying Cyclones over the Atlantic (ERICA) investigated the nature of wintertime over-ocean storms (Hadlock & Kreitzberg, 1988; Lackmann et al., 1997, 1999). This project had similar focus areas as GALE, examining cyclogenesis and the MBL during CAOs and processes involved with coastal frontogenesis. Detailed observations of cyclones during ERICA (Neiman & Shapiro, 1993; Neiman et al., 1993) served as the basis for a new conceptual model (Shapiro & Keyser, 1990) of the life cycle of ETCs that is especially appropriate when cyclones form in confluent flow. The commonly used model up to that time, often referred to as the Norwegian cyclone model (Bjerknes & Solberg, 1922), was shown to be more suitable for diffluent flow conditions (Schultz et al., 1998). That model had been used by synoptic meteorologists to interpret the nature of midlatitude cyclones, but became criticized due to

its inability to adequately explain the range of low-level frontal configurations and observed midlatitude cyclone evolution (Schultz et al., 1998).

Between June 1988 and May 1990, the Eulerian Model Evaluation Field Study (EMEFS) was conducted across the East Coast of North America as part of a joint Canada-U.S. effort to evaluate two Eulerian long-range transport models using surface measurements of air and precipitation chemistry (McNaughton & Vet, 1996). There were also intensive airborne measurements as part of EMEFS that were important to show results such as how O₃ could be simulated well by at least one model (i.e., Acid Deposition and Oxidant Model), but that S, NO₂, and hydrogen peroxide (H₂O₂) were underpredicted aloft (Macdonald et al., 1993).

The Bermuda Atlantic Time-series Study (BATS; <http://bats.bios.edu>) initiated sampling in October 1988 as part of the U.S. Joint Global Ocean Flux Study (JGOFS) program (Lomas et al., 2013). Measurements have been ongoing to the present time at the BATS site ~80 km southeast of Bermuda in the Sargasso Sea. The main thrust of this long-term data record is providing information about processes that drive ocean biogeochemistry on seasonal to decadal time scales, and to better understand how oceans impact the global carbon budget (e.g., Michaels & Knap, 1996; Steinberg et al., 2001). The measurements reported from this site have had important implications for air-sea interactions. A few selected results from an analysis of 10 years of data by Steinberg et al. (2001) include the following: (i) lack of a strong relationship between primary production and particle fluxes, (ii) bacterioplankton are responsible for the majority of living biomass and thus play a key role in carbon uptake, and (iii) the summer is when bacterial biomass, production, and growth rates are more enhanced. Lomas et al. (2013) resolved a number of issues in their analysis of 24 years of data including the reason for the seasonal drawdown of CO₂ when there are no nutrients detectable. They showed that zooplankton are central to carbon removal as they graze and undergo vertical migration. Also, accumulation of dissolved organic carbon (DOC) in the euphotic zone without dissolved organic nitrogen (DON) accumulation in the surface ocean is a contributing factor.

Extensive sea surface measurements have been made over the WNAO, with efforts intensifying in the late 1980s. Many of those early observations focused on model validation of daily latent and sensible heat fluxes using in situ data from coastal field measurement sites as part of the Severe Environment Surface Mooring (SESMOOR), the Coastal Mixing and Optics Experiment (CMO), and the 1993 Acoustic Surface Reverberation Experiment (ASREX93) (Yu et al., 2004 and references therein). Buoy data adjacent to the East Coast have been instrumental in validation of both models and remote sensing products associated with SST (Mesias et al., 2007).

The Atmosphere/Ocean Chemistry Experiment (AEROCE) was a large-scale multi-disciplinary effort to examine biogeochemical cycles over the North Atlantic Ocean as a result of anthropogenic activities with a focus on both marine and atmospheric chemistry (e.g., Arimoto et al., 1995). While significant efforts took place with ground site measurements at Bermuda beginning in 1988, research flights were conducted in 1996 (Milne et al., 2000; Prados et al., 1999). Associated with AEROCE, two SULFIDE cruises in April and October 1989 were conducted as described by Veron et al. (1992), who

demonstrated the utility of lead (Pb) and its isotopes in tracing air masses over the remote WNAO MBL back to continental sources.

Airborne measurements in August – September 1989 were conducted off the U.S. East Coast as part of the first phase of the NASA Global Tropospheric Experiment (GTE) Chemical Instrumentation Test and Evaluation (CITE 3) mission (Anderson et al., 1993; Hoell Jr. et al., 1993; Shipham et al., 1993). The primary goal of CITE 3 was to evaluate airborne instrumentation to measure ambient concentrations of SO₂, hydrogen sulfide (H₂S), carbon disulfide (CS₂), DMS, and carbonyl sulfide (OCS) (Hoell Jr. et al., 1993). Additional objectives included understanding the S cycle and characterizing the impact of continental outflow on O₃ and small aerosol (diameter < 0.5 μm) budgets over the WNAO. Results of CITE 3 included the finding of significant photochemical activity in the MBL of the WNAO when continental air was transported offshore, promoting formation of O₃ and secondary aerosols (Anderson et al., 1993). They also showed that MBL levels of O₃ and small aerosol number concentrations were enhanced by factors of three and six, respectively, when air masses originated over North America versus clean maritime air; the difference in values between the air mass types decreased significantly with altitude. During the CITE 3 study, Shipham et al. (1993) summarized how air masses impacting the WNAO at low levels were governed by the position of Bermuda-Azores High. For instance, maritime tropical air flowed northward along the U.S. East Coast to the WNAO when the anticyclone was based over the central and western Atlantic. As days passed by and the sub-tropical high moved gradually towards the southeast U.S. coast, there was more influence from continental air. At some point later, there was rapid southeastward movement of polar air from Canada containing stratospheric chemical signatures coincident with an intense low over the Canadian Arctic.

3.3 1990 – 2000

A range of measurements including airborne flights took place as part of the North Atlantic Regional Experiment (NARE) along the U.S. East Coast and offshore areas to characterize the evolution of anthropogenic emissions, particularly O₃ and its precursors, in East Coast outflow (e.g., Buhr et al., 1996). Various intensive periods included August – September 1992 (Berkowitz et al., 1995; Doran et al., 1996), August – September 1993 (Fehsenfeld et al., 1996), March – April 1996 (McCaffery et al., 2004), September – October 1997 (Cooper et al., 2001; Fried et al., 2002), and February 1999 (Parrish et al., 2000). Leitch et al. (1996) provided one of the first treatments of ACI for the WNAO, showing that cloud droplet number concentration (N_d) increased as a function of both aerosol concentration and cloud water SO₄²⁻ levels, albeit weakly, and pointed to the need for more measurements.

The first reported measurements of volatile inorganic chlorine gases in the WNAO MBL occurred in January 1992 (Keene et al., 1993; Pszenny et al., 1993). These investigators developed a tandem mist chamber method and quantified concentrations of HCl* (including HCl, ClNO₃, ClNO₂, NOCl) and Cl₂* (including Cl₂ and some HOCl) during a period of onshore flow at Virginia Key near Miami, Florida. Research efforts around Florida continued with the Florida Atmospheric Mercury Study (FAMS), which was a multiyear project between 1992 and 1996 to investigate atmospheric transport and deposition of

mercury (Hg) and other trace metals in central and south Florida (Pollman et al., 1995). Monthly integrated precipitation and weekly integrated aerosol samples were collected at 10 sites around Florida for durations ranging between two to five years (Guentzel et al., 2001). Related to FAMS was the Aquatic Cycling of Mercury in the Everglades (ACME) project aiming to study the hydrological, biological, and geochemical processes governing Hg cycling in the Florida Everglades (Krabbenhoft et al., 1998). Important results from these Hg-based studies in Florida included quantification of wet and dry deposition of Hg around Florida and improving models treating the transport and life cycle of Hg (Pai et al., 1997). Long range transport of reactive gaseous Hg in conjunction with strong convection and thunderstorms during the summer were suggested to account for over half of Hg deposition over southern Florida, with local emissions being less influential (Guentzel et al., 2001). The FAMS data were influential in a later study using tracers of dust to show that dust models could accurately capture seasonal patterns of dust deposition but significantly underestimated summer deposition and yielded latitudinal gradients that were not evident in the observations (Prospero & Landing, 2009).

As a component of University of Maryland's Regional Atmospheric Measurement, Modeling, and Prediction Program (RAMMPP; <https://www.atmos.umd.edu/~rammpp/>), airborne measurements over the mid-Atlantic U.S. began in 1992 (Dickerson et al., 1995; Taubman et al., 2004) with intensive periods during summertime air pollution episodes focusing on aerosol properties and O₃, CO, and SO₂ (Hains et al., 2008; Taubman et al., 2006). Taubman et al. (2004) reported on a series of airborne flights over the U.S. East Coast (Virginia-Maryland) in summer of 2002 with a close look at a smoke plume over a haze plume. Those data coupled to surface and space-borne remote sensing (Vant-Hull et al., 2005) probed the effects of the two layers on both heating rates and atmospheric stability, in addition to highlighting challenges in achieving closure between columnar narrow and broadband shortwave measurements and calculations. Surface measurements were also conducted as part of RAMMPP for the Maryland Aerosol Research and Characterization (MARCH)-Atlantic study; more specifically, gas and aerosol composition were measured at Fort Meade, Maryland during July 1999, October 1999, January 2000, April 2000, and July 2000 (Chen et al., 2003; Chen et al., 2001). One result from these surface observations was that the ratio of elemental carbon (EC) to CO exhibited a temperature dependence, suggestive of EC source strength having been higher in summer periods (Chen et al., 2001).

The Smoke, Clouds and Radiation Experiment (SCAR-A) in July 1993 investigated optical and physicochemical properties of the U.S. East Coast urban plume (e.g., Remer et al., 1997). A series of airborne flights were conducted with one result being that SO₄²⁻ was not close to accounting for the full budget of cloud condensation nuclei (CCN) (Hegg et al., 1995), pressing for more research to identify other components of CCN.

Between July and October 1994, simultaneous measurements of DMS in sea water and in the atmosphere were conducted on the United States Coast Guard heavy icebreaker (USCGC) Polar Sea during a cruise as part of the Canada/U.S. circumnavigation that included data collection over the WNAO and the Sargasso Sea (Sharma et al., 1999). Although the highest atmospheric DMS levels were observed near (south of) the Arctic ice edge, surface water DMS levels were highest in the western Arctic and over the WNAO near

the Sargasso Sea. DMS fluxes varied over several orders of magnitude but were noted to be highest in regions with open water. Further, the atmospheric lifetime of DMS was between 1 to 8 days depending on the location of the measurements, and that modeling results suggested that halogen chemistry (i.e., reactions with Br/BrO) could be an important sink for DMS.

Commercial airliners have been used for atmospheric composition measurements in the upper troposphere/lowermost stratosphere (UTLS) over the WNAO since the 1990s. Ozone and water vapor were characterized using five commercial airliners as part of the Measurements of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program initiated in 1993 (Marengo et al., 1998). Flights were conducted between Europe and North America with one such finding being that peak O₃ levels were in the middle and UT above Germany with the source being North America (Trickl et al., 2003). A subset of flights as part of the Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container (CARIBIC; <http://www.caribic-atmospheric.com/Home.php>) crossed over the WNAO (i.e., the “Caribbean Route”). Results from selected flights between May 2001 and April 2002 afforded insight into the relationship between cloud contact times in the UT and particle number concentrations (Weigelt et al., 2009). After the first phase of CARIBIC (1997 – 2002), a second phase was conducted between 2004 and 2018 with more flights transecting over the WNAO (e.g., Fischbeck et al., 2017). MOZAIC, which had flights up until 2014 (Petzold et al., 2015), and CARIBIC formed the foundation of the operational and sustainable European Research Infrastructure called IAGOS (In-service Aircraft for a Global Observing System; www.iagos.org), which has been providing atmospheric composition measurements in the UTLS since 1994 via tens of thousands of commercial aircraft flights (Petzold et al., 2015). Tables 1 and S1 make reference to those studies from IAGOS making most detailed mention to the WNAO region as defined in this study. An example of the utility of these long-term data is related to findings associated with total odd nitrogen (NO_y), which is the sum of all reactive nitrogen species, in the UTLS. Stratmann et al. (2016) used nine years of CARIBIC data to show there is a strong seasonal cycle for NO_y in both the troposphere and stratosphere. The highest and lowest levels were observed in summer and winter, respectively, for the UT. In contrast, the spring season was characterized by the highest levels for the LS, assisted in part by biomass burning plumes. Gressent et al. (2014) further noted that the high NO_y levels off the U.S. East Coast are linked to NO formation from lightning in conjunction with convective transport to the UT. Cohen et al. (2018) used close to 20 and 12 years of O₃ and CO data, respectively, to show that O₃ has shown a positive concentration trend over time in the UT without a trend in the LS, while the CO trend has been negative in the UT and LS.

To address growing concerns over O₃, the U.S. Environmental Protection Agency (EPA) initiated the North American Research Strategy for Tropospheric Ozone (NARSTO) with the goal of using scientific findings to remediate effects of tropospheric O₃ and to provide air quality policy. In the summer of 1995, the NARSTO-Northeast study was conducted to collect data needed for improved understanding of the regional O₃ issue over the northeastern U.S. As part of that study, Seaman and Michelson (2000) provided a detailed look at a major O₃ event observed in the lower troposphere in mid-July, which was identified as a characteristic high-O₃ event linked to the Bermuda-Azores High, with low winds, high

temperatures, sparse rain, and few clouds over the Northeast. High O₃ from aloft was shown to mix downward, leading to elevated surface levels during the daytime owing to deepening of the mixed layer (Zhang et al., 1998).

The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) mission in July 1996 aimed to reduce uncertainties associated with aerosol effects on climate by examining aerosol properties and effects in the anthropogenic plume over the U.S. East Coast and offshore areas (Russell et al., 1999). Measurement platforms in TARFOX included four aircraft that were coordinated with satellites, surface sites, and ships. TARFOX raised awareness about the significance of carbonaceous constituents in aerosols (Novakov et al., 1997) in addition to hygroscopic growth exceeding what was assumed at that time in climate effect calculations (Kotchenruther et al., 1999). These results were important to address the knowledge gap raised during SCAR-A about SO₄²⁻ not telling the full story about the CCN budget (Hegg et al., 1995). Various forms of closure analyses were conducted with different degrees of success. Hegg et al. (1997) conducted “internal closure” whereby retrieved aerosol optical depth (AOD) was compared to in situ aerosol chemical measurements to show that aerosol extinction was accounted for mostly by water followed by carbonaceous species and then SO₄²⁻. Examples of “external closure” analyses included comparisons of various retrievals of AOD (e.g., surface/ship/airborne sunphotometers, satellite radiometers) (e.g., Durkee et al., 2000; Tanre et al., 1999; Veefkind et al., 1999). Results of such closure analyses were beneficial for validating and improving aerosol models used for direct radiative forcing calculations, which were then compared to values derived from aircraft observations (Hignett et al., 1999). The latter study suggested that more work was needed to quantify impacts of aerosol mixing state on aerosol radiative forcing, which has been a variable problem over time owing to changes in power plant emissions of SO₂ after 2000 (Berg et al., 2016).

The Subsonic Assessment (SSAS) Ozone and NO_x Experiment (SONEX) constituted flights in October – November 1997 over the North Atlantic Ocean to characterize NO_x and O₃ emissions from aircraft (Singh et al., 1999). An important result from SONEX relevant to the upper troposphere was that convection was found to be the major factor leading to the presence of particles and NO_y, and that aviation accounted for at least 7% of the high-altitude particle concentrations (Wang et al., 2000). SONEX overlapped with the Pollution From Aircraft Emissions in the North Atlantic Flight Corridor (POLINAT 2) campaign (September – October 1997), which also had similar objectives to examine the influence of traffic exhaust on upper tropospheric atmospheric composition (Paladino et al., 2000).

Cruises were conducted in May 1998 and September 1999 as part of the Northern Oceans DMS Emissions Model programme (NODEM), which included size-resolved measurements of aerosols to characterize major water-soluble ions and S isotopic composition (Wadleigh, 2004). Results from the latter study showed that SO₄²⁻ contributes to aerosol mass in supermicrometer particles owing to condensation and that methanesulfonate (MSA) is formed in regions with high surface-ocean DMS levels. As will be discussed more in Section 4, the NODEM data revealed higher biogenic contributions to nss SO₄²⁻ than previous reports specifically for Bermuda, with a mean of 35% for submicrometer particles (Wadleigh, 2004).

Around the same time as NODEM, the Aerosols99 cruise from Virginia to South Africa in January –February 1999 focused on measurements of short-lived gas and aerosol species over the Atlantic Ocean (Bates et al., 2001). That cruise experienced seven different ‘air mass’ regimes, with two overlapping with the WNAO (“North America” and “Northern Hemisphere Marine”). The North America air mass exhibited the highest levels for any region of submicrometer SO_4^{2-} and primary organic matter, and supermicrometer NO_3^- , all of which could help explain why chlorine was depleted in sea salt particles (Bates et al., 2001; Quinn et al., 2001).

In February and March of 1999, the Electro-Optical Propagation Assessment in Coastal Environments (EOPACE) campaign took place using the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter (Zielinski & Piskozub, 2005). The focus of EOPACE was to investigate sea salt particle microphysics with dedicated flights in the coastal zone by Duck, North Carolina (Outer Banks). Reid et al. (2001) showed that sizable differences exist in the literature for sea salt size distributions, and that no more than half of salt particle concentration variance is linked to wind speed alone but that other factors need attention such as instrumentation biases, variations in insoluble organic content of the ocean, and natural changes in the ocean’s wind and precipitation fields.

In 1999, a research platform was established at the Chesapeake Light Station (~25 km east of the Virginia coastline) for validation of the Clouds and the Earth’s Radiant Energy System (CERES) and other satellite products (e.g., Rutledge et al., 2006). Seventeen years of continuous radiation measurements were conducted as part of the CERES Ocean Validation Experiment (COVE; <https://cove.larc.nasa.gov/>) until infrastructure challenges stopped measurements in 2016. An example of a COVE outcome is that an ocean albedo look-up table was created to take into account four physical parameters deemed to be influential, including wind speed, ocean chlorophyll concentration, solar zenith angle, and transmission by atmospheric clouds and aerosols (Jin et al., 2004).

3.4 2000 – 2010

The Chesapeake Lighthouse and Aircraft Measurements for Satellites (CLAMS) campaign in July – August of 2001 included airborne, surface, and satellite observations with a goal of validating space-based retrievals of aerosol properties and vertical characteristics of state variables such as temperature and water vapor (e.g., Redemann et al., 2005). The region of analysis was off the U.S. East Coast where flights intercepted days with both clean and polluted air masses (Magi et al., 2005; Reidmiller et al., 2006), categorized into three regimes (Castanho et al., 2005): local pollution/sea salt background, long-range transported dust, long-range transported pollution. Numerous research papers associated with CLAMS were critical in terms of advancing aerosol retrievals over land and ocean for both satellite remote sensors such as the Moderate Resolution Imaging Spectroradiometer (MODIS) (Castanho et al., 2005; Levy et al., 2005; Remer et al., 2005), the Multi-angle Imaging Spectroradiometer (MISR) (Kahn et al., 2005; Reidmiller et al., 2006), and the CERES (Smith et al., 2005), in addition to airborne instruments such as the 14-channel NASA Ames Airborne Tracking Sunphotometer (AATS-14) (Redemann et al., 2005), the Research Scanning Polarimeter (RSP) (Chowdhary et al., 2005), and the Cloud Absorption

Radiometer (CAR) (Gatebe et al., 2005). With the support of MODIS data, in situ observations during CLAMS showed that an incursion of Saharan dust between 24 and 26 July resulted in dust accounting for 40% of fine particulate mass, while on another occasion (17 July 2001) SO_4^{2-} accounted for 70% of fine particulate mass during a regional pollution episode (Castanho et al., 2005).

The Convective Transport of Trace Gases into the Middle and Upper Troposphere over Europe: Budget and Impact on Chemistry (CONTRACE) field experiment in November 2001 collected airborne data and showed that elevated pollution layers over Europe stemmed from North American sources (Huntrieser et al., 2005). Interestingly, the CONTRACE data revealed that enhanced levels of O_3 in such transported plumes were already produced over the Eastern U.S. rather than during transit over the Atlantic Ocean.

As part of a cooperative program between NOAA and the University of New Hampshire, the Atmospheric Investigation, Regional Modeling, Analysis and Prediction (AIRMAP) Institute was established in 2000, which collected surface observational data from sea-level to the top of Mount Washington (~1910 m) to isolate local and regional-scale processes related to air quality in the New England region. While the AIRMAP sites in New Hampshire were operational between 2001 and 2012, their role became especially important during periods of other intensive field campaigns such as those described subsequently, in particular the New England Air Quality Study (NEAQS 2002) (Griffin et al., 2004) and the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT 2004) (Medina et al., 2007). An intriguing finding from AIRMAP efforts was the detection of Asian dust as far east as New Hampshire, Maine, and Massachusetts (DeBell et al., 2004).

Occurring between July and August 2002, NEAQS was comprised of airborne and shipboard measurements around the U.S. East Coast and WNAO (e.g., Bates et al., 2005). The goal was to characterize the sources, transport, and transformation of gases and particles in the MBL off the northeastern coast of the U.S. Selected findings included that $75 \pm 8\%$ of the sub-10 μm dry aerosol mass sampled above the sea surface was smaller than 1 μm , and that the submicrometer aerosol composition was dominated by ammonium sulfate and organic matter (more than $92 \pm 4\%$ of total mass) (Bates et al., 2005). That study's Mie theory calculations offered a result consistent with TARFOX (Hegg et al., 1997), specifically that organic matter was the dominant component contributing to aerosol light scattering, in contrast to long-term monitoring efforts up to that time over the northeastern U.S. that had suggested New England haze was mainly a result of SO_4^{2-} . Certainly the comparisons are impacted by the reduction in SO_4^{2-} over time owing to regulatory activities, which have contributed to organics becoming a more important relative component of the region's aerosols (Bates et al., 2005; Hand et al., 2012a). Measurements directed towards the organic carbon (OC) budget during NEAQS were influential in motivating continued efforts to understand secondary organic aerosol (SOA) formation since it was observed that the increase in submicrometer organic matter in aging air masses off the northeastern U.S. coast could not be explained fully by consumption of what were considered traditional precursor volatile organic compounds (VOCs) at that time (de Gouw et al., 2005). They had suggested

that one explanation could have been that the mechanism for SOA formation was more efficient than assumed.

Leaitch et al. (2010) conducted flights in October 2003 off the Nova Scotia coast as part of the Canadian Surface Ocean-Lower Atmosphere Study (SOLAS). This was an important study as it was perhaps the first airborne effort up until that time to probe ACI. Their results continued what was being learned over time that, rather than SO_4^{2-} , organics are a significant contributor to CCN and thus impact clouds in ways that were poorly understood. The main conclusion of their flights was that the cloud albedo effect caused by anthropogenic carbonaceous aerosols can exceed that of SO_4^{2-} aerosols for the region's stratocumulus clouds. It is noted though that their work was limited by having just two days of flight data, along with the aid of an adiabatic cloud parcel model.

Building off the success of NEAQS, the ICARTT campaign was conducted in July – August 2004 with the goal of continuing examination of regional air quality, long-range transport of pollution, and atmospheric radiative forcing (Fehsenfeld et al., 2006). Within the umbrella of ICARTT, there were efforts such as the Intercontinental Transport of Ozone and Precursors (ITOP) campaign (Lewis et al., 2007), the Chemistry of Halogens on the Isles of Shoals (CHAIOS) campaign (Pszenny et al., 2007; Russell et al., 2007), the Intercontinental Transport and Chemical Evolution Experiment over North America (INTEX-NA) (Singh et al., 2006), a cruise referred to as the New England Air Quality Study (NEAQS) (Quinn et al., 2006; Wolfe et al., 2007), the Intercontinental Transport and Chemical Transformation component of NEAQS (NEAQS-ITCT) (de Gouw et al., 2006), and the Intercontinental Chemical Transport Experiment Ozone Sonde Network Study (IONS) (Thompson et al., 2007a; Thompson et al., 2007b). Note that the NEAQS effort introduced here as part of ICARTT should not be confused with the NEAQS 2002 mentioned earlier and will be referred to here as NEAQS 2004. Table S1 shows that there were over 80 publications from ICARTT, and this number is just limited to those studies having been identified as focusing on the U.S. East Coast and WNAO; several other ICARTT studies were more focused over the continental U.S. such as over the Ohio River Valley. A specific issue discussed in many of the ICARTT reports was biomass burning sources in Alaska and western Canada that impacted the extended WNAO region. Extensive measurements during these studies revealed the importance of considering aerosol and gas transport aloft above the boundary layer rather than only considering surface measurements (e.g., Neuman et al., 2006). Noteworthy were some of the first airborne measurements of nitrate radicals (NO_3) and dinitrogen pentoxide (N_2O_5) yielding results with implications for regional O_3 formation and an underappreciated relationship between NO_x and anthropogenic sulfur emissions (Brown et al., 2006a; Brown et al., 2006b).

Numerous airborne efforts over the WNAO have been rooted in intercomparison with satellite overpass data for validation efforts. Lewis et al. (2010) conducted airborne flights with a lidar over Virginia between 2005 and 2008 and contrasted data with AOD from MODIS. One of their conclusions based on case studies was that AOD could be an indicator of surface $\text{PM}_{2.5}$ levels, especially at high AOD (> 0.3) conditions. Numerous other flights using the NASA High Spectral Resolution Lidar (HSRL) between 2006 and 2014 helped with validation of the Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP)

instrument onboard the Cloud–Aerosol Lidar and Pathfinder Satellite Observations (CALIPSO). While some of these flights were not necessarily associated with a campaign name (2009, 2010, 2014), others were part of either the CALIPSO-CloudSat Validation EXperiment (CC-VEX) in June – August 2006, the CALIPSO and Twilight Zone (CATZ) experiment in July – August 2007, or the Development and Evaluation of satellite ValidatiOn Tools by Experimenters (DEVOTE) mission in September – November 2011 (Kacenenbogen et al., 2011; Painemal et al., 2019; Powell et al., 2009; Rogers et al., 2011; Rogers et al., 2014). Valuable in these comparisons, especially CATZ (McPherson et al., 2010), were ground-based Cimel Sun photometers associated with the NASA Aerosol Robotic Network (AERONET). A notable advancement from these efforts was conducting validation in nighttime lighting conditions (Rogers et al., 2014).

Aspects of air-sea interactions were investigated during two separate wintertime cruises (January 2006, February – March 2007) over the WNAO during the Climate Variability and Predictability (CLIVAR) Mode Water Dynamic Experiment (CLIMODE) (Andersson et al., 2013). In the winter months, periods of cold air that flowed over the Gulf Stream and the North Atlantic subtropical gyre resulted in the greatest amount of heat loss of anywhere in the world. Consequently, the high winds and cooling promoted ocean uptake of carbon dioxide (CO₂), which was the focus of measurements such as those described by Andersson et al. (2013).

Between July and August 2007, the Gulf of Mexico and East Coast Carbon cruise (GOMECC) took place from Galveston, Texas to Boston, Massachusetts. The goal of this cruise was to obtain data on the carbon cycle and related biogeochemical and physical processes in the transition zone between the coastline and open ocean areas. An important result was that oceanic O₃ uptake rate was positively related to wind speed, and the data further suggested that there may be a chemical mechanism in the ocean surface layer that enhanced O₃ uptake to the ocean near land (Helmig et al., 2012). Also, net fluxes of a series of polybrominated compounds were significantly higher for coastal waters in contrast to the open ocean (Liu et al., 2011).

Ship-board measurements in March – April 2008 during the International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) examined the springtime sources, transport, evolution, and impacts of gases and aerosols to and within the Arctic. Several results emerged related to the significant organic fraction of submicrometer aerosols and their functionality (Frossard et al., 2011; Russell et al., 2010).

3.5 2010 – 2019

One of the four parts of the Deriving Information on Surface Conditions from Column and Vertically Resolved Observations to Air Quality (DISCOVER-AQ) flight campaigns was focused on monitoring urban pollution in the Baltimore/Washington D.C. metropolitan region during the month of July 2011. Using multiple aircraft and simultaneous surface observations, the mission was focused on improving interpretation of satellite data to diagnose surface air quality conditions. One study pointed to how the presence of aerosol layers above the boundary layer leads to significant uncertainties in PM_{2.5} estimates based on column-integrated measurements (Crumeyrolle et al., 2014). Chu et al. (2015) further

demonstrated that relying on the synergy of active and passive remote sensing of aerosols can make it more feasible to use satellites for studying air quality on a daily basis. In coordination with DISCOVER-AQ was the Geostationary Coastal and Air Pollution Events-Chesapeake Bay Oceanographic Campaign with DISCOVER-AQ (GEO-CAPE CBODAQ), which was an oceanographic field effort to address topics linked to estuarine biogeochemical processes and air pollution over Chesapeake Bay (Tzortziou et al., 2015). Goldberg et al. (2014) used data from both DISCOVER-AQ and the CBODAQ component to show that O₃ levels were higher over Chesapeake Bay than upwind continental areas, assisted in part by a shallower boundary layer, reduced cloud cover, and slower dry deposition rates.

Related in scope to ICEALOT were summertime cruise measurements conducted in August 2012 between Boston, Massachusetts and Bermuda during the Western Atlantic Climate Study (WACS). Results from WACS yielded insights into primary marine aerosol properties (Keene et al., 2017; Quinn et al., 2014) and organic functional groups and their origin (Frossard et al., 2014; Kawamura et al., 2017). Two years later, the WACS II cruise in May – June 2014 revealed the importance of proteinaceous gels concentrated in the sea surface microlayer that contribute to sea spray aerosols (Aller et al., 2017).

The Two-Column Aerosol Project (TCAP) consisted of one year of intensive surface measurements (June 2012 – June 2013; Cape Cod, Massachusetts) and two intensive periods of airborne measurements in June 2012 and February 2013 (Berg et al., 2016). TCAP aimed to address issues requiring more attention in climate models, especially aerosol mixing state and aerosol radiative forcing. Surface measurements were conducted both at Cape Cod, Massachusetts and several hundred kilometers away over the WNAO (i.e., in different general circulation models (GCM) grid cells) such that the skill of models could be evaluated in simulating horizontal gradients in aerosol microphysical and optical properties. Ground-based and airborne data confirmed what was identified in past studies such as ICARTT that there are widespread aerosol layers aloft that contribute significantly to columnar AOD, with signatures of biomass burning often evident (Berg et al., 2016)

The Ship-Aircraft Bio-Optical Research (SABOR) experiment in July – August 2014 included airborne and ship-based measurements between the U.S. East Coast and Bermuda with the aim of advancing remote sensing capabilities (lidar and polarimetry) for studying ocean biogeochemistry in a region affording a wide range of ocean optical properties (Hair et al., 2016; Ottaviani et al., 2018; Stannnes et al., 2018). One output from SABOR was the reinforcement of the significant potential for scientific advancement of aerosol and cloud microphysics when combining polarimetric and lidar measurements (Ottaviani et al., 2018).

To address the wide discrepancy between field projects conducted over the WNAO in summertime versus other seasons, flights were conducted in February – March 2015 as part of the Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) experiment with a focus on diurnally-resolved composition over the northeastern U.S. Findings included that during wintertime, organic aerosols are abundant with approximately 58% being secondary and originating from pollution sources (Shah et al., 2019). The measured organic species were almost as oxidized during the winter as compared to summer, with wood burning being a key component of primary organic aerosol (Schroder et

al., 2018; Sullivan et al., 2019a). Haskins et al. (2019) investigated the production of molecular chlorine (Cl_2) from the reactive uptake of nitryl chloride (ClNO_2) on aerosol particles during WINTER and showed that this nocturnally important process can yield a non-negligible source of Cl atoms the following morning. While reductions in NO_x and SO_2 emissions have reduced aerosol-phase NO_3^- and SO_4^{2-} , respectively, on the U.S. East Coast during most periods of the year, the wintertime has not experienced such a reduction; to reconcile as to why this is, Shah et al. (2018) showed that this may be attributed to a combination of availability of oxidants and acidity of fine particles (i.e., $\text{PM}_{2.5}$).

The 2015 – 2018 North Atlantic Aerosols and Marine Ecosystems Study (NAAMES) combined ship and aircraft observations focusing on the subarctic portion of the WNAO north of 40°N to study ocean-aerosol-cloud interactions against the backdrop of the annual phytoplankton bloom cycle (Behrenfeld et al., 2019). Quinn et al. (2017) used WNAO cruise data from NAAMES-1, ICEALOT, and WACS II to conclude for the broad region between 70°S and 80°N that the majority of MBL CCN arise from nss SO_4^{2-} aerosols rather than sea salt aerosols. They attributed this to the important influence of entrainment of particles from the free troposphere to the MBL. Sanchez et al. (2018) further explored the seasonal aerosol variability during NAAMES and concluded that changes in the ocean ecosystem and resulting DMS emissions give rise to an increased aerosol SO_4^{2-} contribution to CCN observed during springtime bloom periods, both from new particle formation and from condensation onto existing particles. The NAAMES aerosol-cloud sampling strategy followed a similar set of stacked aircraft legs used by others in ACI-focused campaigns (Crosbie et al., 2016; Sorooshian et al., 2018), and sampled clouds under both typical marine aerosol loadings (order of hundreds per cm^3 of air) as well as severely aerosol-depleted conditions (order of tens per cm^3 of air).

Between September and October 2016, a research cruise with the R/V Endeavor over the WNAO was conducted to investigate marine aerosol production from bursting bubbles and the physicochemical properties of the generated primary marine aerosol (Beaupré et al., 2019; Frossard et al., 2019a; Frossard et al., 2019b). Results from this project have revealed that primarily produced marine aerosols are enriched with marine-derived organic constituents, including surfactants, relative to sea water (Frossard et al., 2019a). Up to 40% of the OC in primary marine aerosol was comprised of refractory dissolved OC (RDOC) and that aerosol emissions by bubble bursting represent an underappreciated removal pathway for old OC from the ocean (Beaupré et al., 2019).

Building off of earlier works over the Chesapeake Bay during GEO-CAPE CBODAQ and other efforts focused on O_3 (Stauffer & Thompson, 2015), including the Hart-Miller Island Pilot Project (Dreessen et al., 2019), the Ozone Water–Land Environmental Transition Study (OWLETS) took place in July – August 2017 (Sullivan et al., 2019b). Airborne, surface, and ship-based measurements over Chesapeake Bay were used to reach the following conclusions: (i) there were significant horizontal and vertical differences in O_3 within just 100 m of land over the water, (ii) O_3 was enhanced over the water especially in the afternoon, and (iii) shipping in the region yielded significant spatial variability in O_3 and NO_2 . The OWLETS 2 project between June and July 2018 was a follow-up effort to

understand the nature of O₃ and related trace gases across the water-land interface near the upper parts of Chesapeake Bay.

A recent airborne campaign between April and May 2018 along the U.S. East Coast, specifically over six old and leak-prone urban centers, studied fugitive methane (CH₄) emissions. Methane is an important greenhouse gas contributing to surface-level O₃ pollution, and has been of major interest in recent years especially over basins producing oil and natural gas. Data collected indicated that CH₄ emission estimates are more than double of what is in the U.S. EPA inventory for the studied regions and are predominantly due to fugitive natural gas losses (Plant et al., 2019).

The Aerosol and Cloud Experiments in Eastern North Atlantic (ACE-ENA) (Wang et al., 2019a) coincided with the NAAMES study time period with deployments of the Department of Energy (DOE) Gulfstream-1 aircraft to the Azores in summer of 2017 and winter of 2018. While ACE-ENA studied ACI in the Eastern North Atlantic, the predominantly westerly flow in this region transports aerosols and trace gases from the WNAO, which can experience significant cloud processing and wet deposition along the way. Thus, the Azores region can provide a complementary perspective of the continued transport and evolution of the North American emissions present in the WNAO. Of note are rich sets of data archived in the Azores at both Graciosa Island (Zheng et al., 2018) and the Pico Mountain Observatory, a free tropospheric site (Honrath et al., 2004; Zhang et al., 2017). Similarly, other ‘bookend’ sites relative to the WNAO region that offer a wealthy inventory of data to contrast results to upwind and downwind regions across the Atlantic Ocean include Barbados (Zuidema et al., 2019), the Canary Islands (Maring et al., 2000), Cape Verde (Carpenter et al., 2010), and Puerto Rico (Reid et al., 2003b).

4. History of Bermuda and Sargasso Sea Research

A subset of the field studies described up until now have included some degree of attention towards Bermuda, but here we focus on those studies that were more intimately linked to the island and the adjacent Sargasso Sea (Figure 1). Research activities have been intense at Bermuda over past decades as it represents a site ideal for long-term data relevant to climate and atmospheric changes (e.g., Aryal et al., 2014). The long-term nature of measurements available at Bermuda has allowed for specific lines of inquiry that would not be possible with short-term datasets. Just as one example, Galloway et al. (1993) used continuous data of aerosol and precipitation composition over a 2-yr period to more robustly quantify scavenging ratios.

Similar to Section 3, Tables 1 and S1 show a breakdown of the categories of Bermuda and Sargasso Sea research identified through an extensive literature survey. Note that individual studies could have covered multiple categories. Many of the studies were associated with AEROCE, which was already introduced in Section 3. Among the 146 peer-reviewed studies classified as Bermuda research, the majority were devoted to investigating aerosol properties (59), followed by gases (38), and wet deposition (37), with the overwhelming majority of these focused on chemical composition and source apportionment. The least examined topic

was ACI with just four publications. A brief overview of the research results is provided below subsequent to a brief summary of the island characteristics and air flow climatology.

4.1 Island Characteristics and Air Flow Climatology

Bermuda is a British Overseas Territory 30 km long and 2 km wide, approximately 1000 km east of Cape Hatteras of North Carolina, U.S. (Figure 1). This island had a population of ~64,000 as of 2016 (Government of Bermuda, 2019) with extensive tourism. Signatures of local pollution have been reported to be significantly outweighed by transported pollutants (Galloway et al., 1988). The relative importance of anthropogenic versus biogenic sources varies based on season and transport pathway (Arimoto et al., 1992; Galloway & Whelpdale, 1987; Huang et al., 1996). Results from AEROCE showed that pollutants arriving at Bermuda exhibited strong seasonal cycles, such as with dust and sea salt, and that the influence of North American pollution at Bermuda varied seasonally with most influence in wintertime (Anderson et al., 1996; Arimoto et al., 1995; Arimoto et al., 2003; Galloway et al., 1989; Moody et al., 1995; Wolff et al., 1986).

The island's flow climatology is governed by the famous anticyclone that bears its name. The months of May – November are dominated by flow around the subtropical anticyclone, while westerly-northwesterly flow is more common in the remaining cooler months between December – April (Merrill, 1994). In their analysis of seven years (January 1975 – December 1981) of airmass back-trajectory data, Miller and Harris (1985) showed that 60% of the time Bermuda experienced direct flow from North America, while southeasterly and southwesterly flow were especially more prominent during high rain periods, and thus influenced the overall chemistry more during those times. Precipitation at Bermuda is largely governed by frontal and convective rains (Galloway et al., 1993). A review of subtropical storms near Bermuda from 1957 – 2005 found that most passed near the island in September and October (Guishard et al., 2007). The direct passage of Hurricane Fabian over Bermuda in 2003 was used to study the impacts of topography on surface winds and subsequent structure damage (Miller et al., 2013).

4.2 Pollution Characteristics

Various methods of analysis have been used for air mass source apportionment including some combination of the following (e.g., Keene et al., 2014): (i) chemical measurements (especially ratios) in gas, aerosol, and precipitation (e.g., Chen & Duce, 1983; Cutter, 1993; Savoie et al., 2002), (ii) radionuclides (Arimoto et al., 1999), (iii) trajectory modeling (e.g., Merrill, 1994; Merrill et al., 1996), (iv) isotopic measurements (Hastings et al., 2003; Lin et al., 2012; Turekian et al., 2001), and (v) remote sensing (Rogers et al., 2019). Most studies for the island suggest that anthropogenic sources over North America are the dominant contributor of pollution over Bermuda (Dickerson et al., 1995; Keene et al., 2014). Early measurements (May 1980 – April 1981) directly linked North American outflow to acid rain on Bermuda, with sulfuric acid (H_2SO_4) being almost wholly responsible with very minor HNO_3 contributions (Jickells et al., 1982). However, more recent evidence suggests that owing to regulatory activities in North America (e.g., Clean Air Act, ban on leaded gasoline) and Europe, the relative influence of anthropogenic sources at Bermuda may have changed (Keene et al., 2015; Keene et al., 2014). Recent years reveal decreasing trends in AOD

(Streets et al., 2006), pollutants in the gas phase such as O₃ and SO₂ (Hand et al., 2012a; Kumar et al., 2013; 2018; Vestreng et al., 2007), particulate mass concentrations (Hand et al., 2012a; U.S. Environmental Protection Agency, 2018), and SO₄²⁻ and NO₃⁻ concentrations in wet deposition (Leibensperger et al., 2012a). Even with a decreasing trend in aerosols and potential atmospheric cooling over the WNAO since the early 1990s, Moody et al. (2014) showed that the impact of aerosol transport from the northeastern U.S. is still very significant at Bermuda, as it accounted for direct radiative cooling between 1.0 – 2.2 W m⁻² at the time of that study.

Many species have been characterized in the aerosol and precipitation phases. Common species studied in aerosol and precipitation samples include numerous trace elements (Anderson et al., 1996; Arimoto et al., 1995; Arimoto et al., 1992; Cutter, 1993; Ellis et al., 1993), inorganic species (Moody & Galloway, 1988; Savoie et al., 2002), organic acids (Keene et al., 2015; Turekian et al., 2003), total nitrogen, and isotopic composition of species such as ammonium (Altieri et al., 2016; Altieri et al., 2013; Altieri et al., 2014). Additional constituents measured in the aerosol phase included radon (Allen et al., 1996; Colle et al., 1995; Hutter et al., 1995), radionuclides such as ²¹⁰Pb and ⁷Be (Arimoto et al., 1999), and plant-based leaf wax biomarkers (Conte & Weber, 2002a).

A few aerosol studies are worth noting that examined issues other than reporting chemical composition and sources. Aerosol scattering was characterized by Aryal et al. (2014), showing that columnar optical properties are weakly related to surface optical properties, motivating the importance of vertically-resolved measurements. The acidity of regional aerosols was characterized at Bermuda in multiple studies (Keene et al., 2002a; Keene & Savoie, 1998); Keene et al. (2002a) showed that supermicrometer particles, mainly comprised of sea salt, exhibited pHs in the upper 3s and 4s, whereas finer aerosols had pHs in the 1s and 2s. Several studies based in Bermuda laid the foundation of knowledge in how to apply caution when quantifying various parameters associated with sea salt and non-sea salt constituents (e.g., Keene et al., 1986). In their study of the effects of wet deposition on optical characteristics of ambient air at Bermuda, Todd et al. (2003) highlighted the complex interaction between meteorology, radiative transfer, and atmospheric chemistry en route to showing that precipitation removes optically active aerosols. Years earlier, Galloway et al. (1993) concluded that near-surface Na⁺ was most efficiently scavenged by rain while nss SO₄²⁻ was least effectively scavenged. However, they investigated only a limited number of species including NO₃⁻ and MSA.

Gases characterized over Bermuda have been wide-ranging with most examining O₃ (Boylan et al., 2015; Cooper et al., 1998; Oltmans & Levy, 1992, 1994), but also OCS (Berkelhammer et al., 2016), methane (CH₄) (Bergamaschi et al., 2009; Turner et al., 2016), CO₂ (Chevallier et al., 2010), CO (Petron et al., 2002), NO_x (Saikawa et al., 2014), SO_x (Wolff et al., 1986), cyclic siloxanes (MacLeod et al., 2013), sulfur hexafluoride (SF₆) (Rigby et al., 2010), polychlorinated dibenzo-p-dioxins and dibenzofurans (Baker & Hites, 1999), water vapor (Benetti et al., 2017), per- and polyfluoroalkyl substances (PFAS) (Gawor et al., 2014), and various other persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) (Genualdi et al., 2010; Panshin & Hites, 1994),

polychlorinated naphthalenes (PCNs) (Lee et al., 2007), and organochlorine pesticide species (Harner et al., 2006; Koblizkova et al., 2012a).

Some selected results related to O₃ are worth highlighting. In their study of transport climatology of O₃ over Bermuda, Moody et al. (1995) showed a significant contribution from the upper troposphere and lower stratosphere. That work confirmed earlier results showing that enhanced surface O₃ concentrations were due to large-scale subsidence from the mid-troposphere (Oltmans & Levy, 1992). Airborne measurements during AEROCE indicated that one such source of the mid-tropospheric O₃ was upstream frontal systems that inject dry O₃-rich air aloft (Prados et al., 1999). Other work showed that O₃ increased with altitude under postfrontal conditions in contrast to a more homogenous profile under prefrontal conditions (Cooper et al., 1998). Merrill et al. (1996) pointed to the importance of active planetary wave breaking in promoting stratosphere-troposphere exchange over the WNAO leading to enhanced O₃ at Bermuda in the summertime (Dickerson et al., 1995).

4.3 Isotopic Studies

There has been an impressive amount of isotopic research at Bermuda offering insight into a range of processes (Gobel et al., 2013; Hastings et al., 2003; Knapp et al., 2010). Total aerosol SO₄²⁻ has been apportioned into different contributors (sea salt, anthropogenic, marine biogenic) based on sulfur isotope data (Lin et al., 2012; Turekian et al., 2001). Turekian et al. (2001) used detailed size-resolved measurement to report that marine biogenic sources contributed only 15 ± 7% and 11 ± 2% to total supermicrometer and submicrometer nss SO₄²⁻, respectively, over Bermuda. The subsequent study by Lin et al. (2012) showed that over the central North Atlantic, 50–80% of submicrometer SO₄²⁻ was of marine biogenic origin, in contrast to near the coast of North America where most aerosol SO₄²⁻ was of anthropogenic origin. For the sake of comparison to these two Bermuda-related studies, S isotopic composition data from NODEM (Section 3) indicated that biogenic contributions to nss SO₄²⁻ in submicrometer particles ranged from 4% to 65% (mean = 35%) (Wadleigh, 2004). Although to the east of our defined WNAO domain, Patris et al. (2000) used S isotopic composition data over the Atlantic in the Northern Hemisphere to show that 50–90% of nss SO₄²⁻ was biogenic for coarse particles (diameter > 2.7 μm), with the biogenic contribution to nss SO₄²⁻ being less than 35% for smaller particles. It is unclear exactly as to what factors can explain the differences between these various studies.

Isotopic composition measurements for OC and oxalate in size-segregated aerosols helped show that supermicrometer particles had a significant enrichment in marine-derived OC relative to seawater (724 times higher) (Turekian et al., 2003). Lightning was noted to be a significant source of NO₃⁻ over Bermuda during the warmer months of April through September based on nitrogen and oxygen isotopic composition measurements for NO₃⁻ in rain (Hastings et al., 2003). Source apportionment of aerosol iron (Fe) was conducted using Fe isotope analysis, with a conclusion being that biomass burning may be a likely source of Fe in the fine size fraction (< 2.5 μm) rather than soil or other sources (Mead et al., 2013). Isotopic chlorine measurements at Bermuda showed that HCl evaporation was the primary route of chlorine volatilization from aerosols (Volpe & Spivack, 1994). Isotopic Pb research has also been an area of interest over the Sargasso Sea, confirming that when leaded gasoline

was phased out in North America and Western Europe, simultaneous reductions were observed in Pb levels over the WNAO (Veron et al., 1998; Veron et al., 1993). Chen et al. (2009) showed that anthropogenic activity has contaminated surface ocean water and the global geochemical cycle of osmium (Os) based on Os isotopic measurements. Bonne et al. (2015) used water vapor isotope measurements to trace an atmospheric river event from the western subtropical North Atlantic Ocean to Greenland. Altieri et al. (2016) used N isotopic measurements in rain and aerosols to conclude that marine biogenic emissions are responsible for water-soluble organic N (WSO_N) and that in-cloud scavenging (rather than below-cloud scavenging) accounts for the majority of WSO_N in rain. Based on measurements of leaf wax compounds in Bermuda aerosols, it was shown that $\delta^{13}\text{C}$ coincided with the seasonal cycle of midlatitudinal terrestrial productivity (Conte & Weber, 2002b).

4.4 Sargasso Sea Research

Associated with research at Bermuda have been numerous investigations examining various aspects of air-sea interactions and pollutant deposition over the Sargasso Sea, many of which have been linked to the BATS program. One subset of air-sea interaction research near Bermuda has focused on the impacts of hurricanes (Babin et al., 2004; Black & Dickey, 2008; Dickey et al., 1998; Koch et al., 2009; Nelson, 1998; Zedler et al., 2002) and winter storms (Kim et al., 2019; Lomas et al., 2009) on oceanic properties, structure, and composition, including impacts on phytoplankton.

Multiple studies have focused on the impacts of atmospheric aerosols at the air-sea interface. Seminal works by Hoffman and Duce (1976, 1977) pushed forward understanding of factors leading to OC enrichment in sea salt particles, including the amount of surfactants and nature of organic constituents in seawater, and the distance bubbles traveled in seawater during bursting at the air-water interface. Measurements in the upper 200 m of the Sargasso Sea water column linked aeolian aerosol deposition to the presence of various crustal elements such as aluminium (Al) (Jickells et al., 1990). Wet deposition was also shown to be a major route for the aeolian flux of cobalt (Co) (Shelley et al., 2012). Subsequent works extended the effort to characterize the degree of atmospheric inputs of various trace elements (Jickells et al., 1994; Jickells et al., 1998; Kim et al., 1999; Tian et al., 2008) and radionuclides (Kadko et al., 2015; Kadko & Prospero, 2011) to the Sargasso Sea. Other works have shown that the input of various metals (Mackey et al., 2012a; Mackey et al., 2012b) and inorganics such as NO_3^- and ammonia (NH_3) (Michaels et al., 1993) promoted productivity of phytoplankton communities. Orcutt et al. (2001) quantified N_2 fixation rates by *Trichodesmium* based on monthly cruises around the BATS site in the Sargasso Sea; those results provided a link between aeolian dust and Fe flux to the sea. The effect of sea water properties (temperature, pH, deoxygenation) on aerosol Fe dissolution was examined showing that ocean warming and acidification is unlikely to be as influential in Fe dissolution in contrast to changes in land use and fossil fuel combustion (Fishwick et al., 2014).

The role of sea spray in transporting nonylphenols (NPs) and nonylphenol ethoxylates (NPEOs), which are surfactants relevant at the air-water interface, was investigated with a

combined laboratory, field, and modeling effort (McInnes et al., 2013). They showed that aqueous aerosols are enriched with NPs and NPEOs as compared to bulk source water, motivating the need for models to consider spray generation and the potential for subsequent droplet-gas exchange to accurately simulate the fate of such surfactants in marine aerosols.

4.5 Development and Validation Studies

A large number of instruments, techniques, and models have been demonstrated and/or validated based on research at Bermuda, in part assisted by its unique positioning in the Atlantic Ocean. Maring and Schwartz (1994) successfully deployed modified versions of commercial clean room condensation particle counters for continuous measurements at Tudor Hill, one of the most used research facilities on the island, over a year with just minor maintenance. As part of the Global Atmospheric Passive Sampling (GAPS) network, extensive efforts over Bermuda in unison with other global sites demonstrated the utility of passive samplers to provide a spatial view of POPs (Koblizkova et al., 2012b; Pozo et al., 2009; Pozo et al., 2006). Sholkovitz and Sedwick (2006) deployed a buoy-mounted aerosol sampler on the Bermuda Testbed Mooring as part of their investigation of Fe and sea salt characteristics. AERONET data collected at Bermuda were helpful in developing a maritime look-up table kernel for retrieval algorithms and as a source of input to aerosol transport models and radiative forcing calculations (Smirnov et al., 2003). Tomza et al. (2001) explored the use of color on particulate sample substrates to infer information about aerosol composition. They linked brown and gray to mineral dust and absorbing aerosol, respectively, with white indicating the absence of the latter two aerosol types.

5. Synthesis of Results and Future Outlook

An extensive historical record of research was provided above and this section aims to distill selected key findings and future research needed around seven research themes used to categorize publications in Tables 1 and S1. We note that the eighth category (i.e., meteorology and air flow analysis) is already covered adequately in Section 2 and thus is not discussed further here. Furthermore, the category of wet deposition is now expanded to include dry deposition in Section 5.3.

Continued research efforts are critical for a number of reasons. First, improvements in measurement capabilities and models and opportunities for sustained long-term observations from airborne platforms provide a pathway forward to gain new insights (Sorooshian et al., 2019). Second, past works reviewed in this paper have raised new questions that warrant investigation. Third, there have been changes over time in this region that allow for an opportunity to contrast conditions between periods separated by decades; for instance, as previously noted, the overall level of pollution has decreased over recent decades (Robson et al., 2018; Yoon et al., 2014; Zhang & Reid, 2010; Zhao et al., 2008) and so has the general profile of gas, aerosol, and precipitation composition (Feng et al., 2019; Hand et al., 2012a; Keene et al., 2015; Robson et al., 2018), which may have ramifications for clouds, precipitation, and radiative transfer in the region.

5.1 Gases

The timeline of work in Sections 3–4 demonstrates that significant advancements have been made in the fundamental understanding of atmospheric chemistry related to anthropogenic and marine emissions transmitted through (and emitted from) the WNAO region. Improvements have also been made in quantifying the impacts of pollution emitted from the WNAO region on downwind regions such as Bermuda and Europe. A common theme among many studies has been the clear evidence of the success of regulatory activities in reducing concentrations of anthropogenic pollutants. More specifically, there have been significant reductions in NO_x , SO_2 , and O_3 precursor emissions from continental regions upwind of the WNAO region in the last 30 years (Feng et al., 2019; Frost et al., 2006; Robson et al., 2018).

Here we add to the growing collection of data products showing decreasing trends linked to anthropogenic pollution over the East Coast of North America and extending over the WNAO region. In contrast to most data records used to point to reductions in pollutant levels that are based on surface monitoring data, satellite data offer the advantage of broader spatiotemporal coverage. One major pollutant that has shown a decrease in emissions is CO. Columnar CO concentrations were obtained from the Atmospheric Infrared Sounder (AIRS) onboard the NASA Aqua satellite. For each day between 2003 and 2018, a subset of gridded data pixels falling into the domain bounded by 25 – 50°N and 60 – 85°W were selected. These daily maps of data were then averaged seasonally for two time spans: 2003 – 2010 and 2011 – 2018 (Figure 4). The spatial profile of CO is indicative of pollution outflow from the East Coast to the WNAO region, with a domain-averaged maximum value in March – April – May (MAM) and a minimum in JJA. Reductions in CO are evident in the most recent set of years, with areas of greatest decrease varying based on season. In terms of domain-wide average values, the percent reduction in CO from 2003 – 2010 to 2011 – 2018 for each season was as follows: December – January – February (DJF) = 5%, MAM = 7%, JJA = 3%, September – October – November (SON) = 3%. As AIRS data are columnar in nature, it remains to be learned what the relative influence is of air masses aloft enriched with CO (such as with biomass burning plumes) in contrast to boundary layer air on both seasonal and interannual scales.

While concentrations of anthropogenic gases such as CO over the extended WNAO region have decreased over time (Figure 4), biogenic gases are less sensitive to regulatory activities. One such gas with both biogenic and anthropogenic sources that has been investigated over the WNAO region is acetone (Fischbeck et al., 2017), which is an important source for hydroxyl radicals (HO_x) and O_3 (Singh et al., 1995). Under high- NO_x conditions, acetone can promote formation of peroxyacetyl nitrate (PAN), which itself can act as a temporary NO_x reservoir (Fischer et al., 2014). An important marine biogenic gas, DMS, exhibited a strong anticorrelation with the NO_3 radical due to their reaction with each other suggesting that DMS oxidation by NO_3 may have an underappreciated impact on SO_4^{2-} formation in the WNAO MBL (Stark et al., 2007). The overall impact of DMS and other marine biogenic gases are an area requiring more research over the WNAO to understand how they impact both gas and aerosol budgets. Biomass burning emissions also can significantly impact gas budgets over the extended WNAO region, and more measurements are warranted to further

understanding and follow up on successful past missions examining long-range transported gas emissions from Alaska and Canada wildfires (Warneke et al., 2006), but also with consideration of transport paths of biomass burning plumes from Mexico and the Southeast U.S.

Ozone has been a major pollutant of concern in past research for the extended WNAO region including both upwind and downwind regions. The aims of past works examining O₃ have largely been studying O₃ precursor emissions (VOCs, NO_y, NO_x, and SO₂), identifying previously unknown precursors and sinks (HNO₃, NO₃, and N₂O₅) in the surface O₃ reaction chain (Fehsenfeld et al., 2006; Fischbeck et al., 2017; Gressent et al., 2014; Petzold et al., 2015), and identifying sources and concentrations of halocarbons (Zhou et al., 2005) that have been implicated in depletion of stratospheric O₃ (Sommariva et al., 2011). Surface O₃ exhibits a strong seasonal cycle over the WNAO, including maximum and minimum values in spring and summer, respectively (Robson et al., 2018). While surface concentrations have decreased over time at Bermuda likely linked to reductions in precursor emissions over North America (Granier et al., 2011), columnar values have exhibited an increase (in addition to CH₄) between 2005 and 2016 (Robson et al., 2018), reflective of other influences such as long-range transport and variability in temperature and solar radiation. Differences exist between studies examining O₃ trends over time, partly due to differences in measurements and retrieval algorithms (Ebojje et al., 2016; Oetjen et al., 2016; Parrish et al., 2014). Parrish et al. (2014) pointed out that the current understanding of tropospheric O₃, or rather the inclusion of chemistry and transport into chemical climate models, is still incomplete, which leads to disagreements still between models and measurements in northern midlatitudes. Therefore, research is certainly needed to continue unraveling details controlling the horizontal, vertical, and temporal variations of O₃ over the WNAO region, including looking at the impact of intercontinental transport, lightning generation of NO_x, vertical transport processes, and NAO phase (Dickerson et al., 1995; Gressent et al., 2014; Huntrieser et al., 2005; Neuman et al., 2006).

Nitric acid (HNO₃) is a particularly important gas-phase species over the extended WNAO region due to widespread sources of its precursor (NO_x) across the East Coast (Fischer et al., 2006; Jordan et al., 2000). HNO₃ is involved with various processes impacting aerosol composition such as acid-base chemistry with NH₃ to form ammonium nitrate salts or partitioning to coarse aerosol surfaces such as sea salt and dust; furthermore, it is known to partition very effectively in cloud droplets (Hayden et al., 2008; Prabhakar et al., 2014). A series of works investigated the interplay between N₂O₅, NO_x, and HNO₃, which influences O₃ and VOC concentrations (Aldener et al., 2006; Brown et al., 2006b). Brown et al. (2006a) identified that the uptake coefficient of N₂O₅ on aerosol particles depends significantly on aerosol composition, especially SO₄²⁻ amount. Another important impact of HNO₃ (and H₂SO₄ and organic acids) in the WNAO region is to liberate chloride from sea salt particles (Finlayson-Pitts & Pitts, 2000; Graedel & Keene, 1995), which subsequently alters oxidant budgets and atmospheric chemical reactions.

Building off the well-documented topic of chloride depletion, halogen radical chemistry has been an active area of research with still major uncertainties for the WNAO region. This is an important issue because with enhanced emissions of acids and their precursors in areas

like the East Coast of North America with large populations and anthropogenic activity, there is a high likelihood of acid-catalyzed halogen activation and cycling with major implications for atmospheric chemistry and climate (Long et al., 2014a; Sander et al., 2003). A modeling study from CHAiOS showed that as continental air advects over the ocean, significant acid displacement occurs on fresh sea salt particles, leading to high Cl_2 levels after oxidation of HCl by OH (Pechtl & von Glasow, 2007). When that air re-circulates back to the coast via onshore winds, high Cl_2 levels results at nighttime. Other results from CHAiOS indicated that Cl reactions boost the reactivity of VOCs by up to 30% over that due to OH alone (Pszenny et al., 2007). As noted already, WINTER results showed that the reactive uptake of ClNO_2 on aerosol particles is an important source of Cl_2 during nocturnal periods (Haskins et al., 2019). When including an advanced representation of tropospheric Cl chemistry into the GEOS-Chem model, Wang et al. (2019b) showed that the model could reproduce vertical profiles of HCl from the WINTER campaign, in addition to ClNO_2 being mainly concentrated in the MBL at night. A deficiency was that the model overpredicted Cl_2 at night, which was suggested to be linked to uncertainties in the rate of the heterogeneous reaction between ClNO_2 and Cl^- .

In addition to Cl, Br is also an important constituent of sea salt that can be significantly depleted in supermicrometer particles with resultant inorganic reactive gases (e.g., BrCl) subsequently impacting O_3 , DMS, hydrocarbons, and various other marine constituents (Sander et al., 2003). Long et al. (2014a) examined how tropospheric chemical composition is sensitive to halogen radical chemistry (Cl, Br, I, NO_x) with model simulations compared to observational data. Although their study was global in nature, the following selected salient points are relevant to the WNAO in terms of areas requiring more attention: (i) cycling of sea salt Br is important for the free troposphere where Br cycling is closely linked to MBL chemistry, (ii) the spatial distribution, partitioning, and impacts of inorganic Br species are influenced strongly by large-scale circulation, (iii) cycling of Br from marine aerosol is influenced greatly by marine aerosol fluxes, and (iv) net production of BrCl in the troposphere may be too efficient in models (Keene et al., 2009; Sommariva & von Glasow, 2012) as volatile inorganic Br mixing ratios are overpredicted. As marine aerosol particles are enriched with organics including in their surface layers (Kolb et al., 2010; Long et al., 2011), work is needed to evaluate the impact of these organics on inorganic halogen cycling including reactions with Cl and Br radicals (Long et al., 2014a; Sommariva & von Glasow, 2012). Research to address speciation of a broader range of Br-containing species in the MBL is encouraged, in addition to gathering statistics on the diurnal and seasonal variations in the Cl and Br enrichment factors (i.e., ratio of Cl or Br to Na in a sample relative to pure seawater) and their vertical MBL profiles (Sander et al., 2003). As demonstrated recently for the Southeast U.S. (Zhu et al., 2019), gaining a better understanding of spatiotemporal patterns of lightning can have important implications for simulating NO_x profiles, and thus their overall impact on atmospheric chemistry and generated biases in satellite retrievals of NO_2 .

5.2 Aerosols

The topic given most attention in past research over the WNAO has been aerosols, ranging from characterization of their chemical, optical, and size properties to understanding their

sources and aging behavior. Similar to anthropogenic trace gases such as CO, the surface aerosol burden has decreased considerably over the Eastern U.S. (primarily from controls on the sources of primary and secondary SO_4^{2-}) since peaking in the 1970s – 1980s (Leibensperger et al., 2012a, 2012b; Streets et al., 2006; Yang et al., 2018). Trend analysis of satellite-based observations of AOD suggest that this decrease also extends over the WNAO (Colarco et al., 2014), which may alter the relative contributions and composition of transported versus locally-produced aerosols in the WNAO atmosphere. Consequently, new observations are needed to constrain the current WNAO aerosol and CCN budget, and to determine to what extent the long WNAO data record still applies to the present day.

As a way to summarize the current spatiotemporal view of aerosols in the extended WNAO region, seasonal profiles of AOD are shown for data between 2014 – 2018 (Figure 5). The WNAO region exhibits a characteristic AOD annual cycle (e.g., Hsu et al., 2012), with high AOD in summer especially over the subtropical zonal band south of 25°N (e.g., Saharan dust) and U.S. East Coast. The decline in AOD moving east of the North America coast is representative of pollution outflow. Minimum AOD values occur in winter, with a zone of higher values north of 35°N attributed to primary marine aerosols such as sea salt. The AOD annual cycle over the continental U.S. is consistent with ground-based observations of $\text{PM}_{2.5}$ (Hand et al., 2012b), with high concentrations in summer likely attributed to aerosol stagnation and photochemical processes (Tai et al., 2010).

Key questions remain as to the role of marine aerosol particles, including primarily emitted particles and secondarily produced species via gas-to-particle conversion processes. It is well known that ocean ecosystems emit DMS gas that subsequently oxidizes to form either methanesulfonic acid (MSA) or SO_2 , the latter of which can eventually form particulate SO_4^{2-} . SOA can also be formed in the MBL due to emissions of marine biogenic VOCs, including end products such as alkyl amines (Facchini et al., 2008a; Youn et al., 2015) among others. It has also been hypothesized that increased organic compounds in the seawater translate into elevated organic aerosol emissions via bubble-bursting processes. The photic zone contains dissolved organic matter comprised of carbohydrates and particles containing viruses, bacteria, algae, and organic detritus that can bind together by exopolymer gels and contribute to the MBL aerosol budget after bubble bursting (Bigg et al., 2004; Facchini et al., 2008b; Quinn & Bates, 2011). A large body of work for the WNAO region, including over the Sargasso Sea, provides consensus that primary marine aerosols ejected from the sea surface are highly enriched in marine-derived organics relative to sea water (Beaupré et al., 2019; Frossard et al., 2019a; Frossard et al., 2019b; Keene et al., 2007a; Long et al., 2014b; Quinn et al., 2014), and that these organic-laden aerosols are both CCN-active (Quinn et al., 2014) and reactive (Zhou et al., 2008).

Understanding the role of clouds and humidity in impacting SOA formation and evolution has received scarce attention over the WNAO, in contrast to other regions such as the northeastern Pacific Ocean (Sorooshian et al., 2010; Sorooshian et al., 2013). Schum et al. (2018) showed that the oxygen-to-carbon (O:C) ratio for wildfire-influenced aerosols ($\sim 0.45 - 0.48$) transported over 7 – 10 days in the free troposphere to the PICO Mountain Observatory in the North Atlantic was lower compared to that of North American outflow aerosols transported mainly through the MBL over only three days (~ 0.57). While the O:C

ratio difference was attributed to the free tropospheric particles being more viscous and thus less vulnerable to oxidation than the MBL aerosols, it is worth considering in future model-measurement studies how enhanced humidity and cloudiness in the WNAO MBL could enhance the O:C ratio via aqueous oxidation pathways (Ervens et al., 2018; Ervens et al., 2014). Further, it is unclear as to what the vertical and spatial profile is of brown carbon over the WNAO and how those profiles are impacted by humidity and clouds.

More attention is warranted towards the spatiotemporal profiles of aerosol size distributions, and how they are impacted by meteorology, boundary layer dynamics, and air mass trajectories. Related to the coarse end of particle size distributions, the underappreciated role of giant CCN in affecting cloud properties should be addressed with more concerted efforts to better constrain their horizontal and vertical profiles in the WNAO MBL such as what was attempted for the North Carolina Outer Banks region (Reid et al., 2001) and in other marine regions (Colon-Robles et al., 2006; Reid et al., 2006). A recent study discussing wintertime measurements of fine particles over the U.S. East Coast (Raleigh, North Carolina) demonstrated the importance of considering boundary layer evolution to understand sources and transport of fine particles (Meskhidze et al., 2019). More specifically, the break-up of the nocturnal inversion as days progressed helped bring particles from aloft in the residual layer to the surface where concentrations became comparable to those earlier during rush hour periods.

Another unresolved issue is the discrepancy in the annual cycle magnitude between satellite AOD and ground-based surface mass observations (Goldstein et al., 2009). While that study hypothesized that biogenic organic aerosols could account for the dominant peak in AOD during the summertime, analysis of aerosol extinction coefficient profiles from CALIOP suggested another factor being aerosol production above the surface (Ford & Heald, 2013). This raises an important issue warranting more aggressive research, which is also central to the limitations of AOD analysis such as in Figure 5 and several other remote sensing studies (Jongeward et al., 2016; Zhao et al., 2008) and surface-based efforts (Hand et al., 2012a; Hand et al., 2012b). In particular, vertically-resolved measurements are needed with broader spatial and temporal coverage than what is typically offered by short-term intensive periods that often occur in summertime (see Section 3). Two especially important aerosol types that can be transported aloft in the free troposphere and have received considerable attention over the WNAO include dust and biomass burning emissions. They are each discussed in more detail below.

5.2.1 Dust—North African dust reaches the WNAO every summer, though measured in Bermuda in lower quantities and for a shorter season than in Miami or Barbados (Arimoto, 2001; Muhs et al., 2012; Prospero, 1999; Prospero & Landing, 2009). Dust amounts collected at Bermuda indicate a clear latitudinal gradient from those sampled at Barbados and Miami (Muhs et al., 2012), and a July – August maximum that is slightly shorter in duration than at the two other locations, particularly Barbados. Dust sampling at Bermuda was undertaken as part of the AEROCE program, and included a focus on determining the trace elements associated with the dust (Arimoto et al., 1995; Arimoto et al., 2003; Arimoto et al., 1992; Sholkovitz et al., 1993). The accumulation of dust through long-range transport from northern Africa, as opposed to North America, over ~500 ka years is held responsible

for the ruddy character of Bermuda soils (Muhs et al., 2012). This phenomenon of dust transport and deposition is significant for a variety of reasons outlined in previous works: (i) contribution to regional soils (Trapp et al., 2010), (ii) source of nutrition for oceans owing to constituents in dust that may be limiting micronutrients such as Fe, phosphorus (P), and nitrogen species (Duce et al., 2008; Mahowald et al., 2008), (iii) impact on regional climate and clouds (IPCC, 2013), and (iv) ability to transport microbes (Prospero et al., 2005).

Many studies have examined dust transport to the WNAO and Caribbean regions with satellite and reanalysis data, ground observations, and transport modeling (e.g., Chen et al., 2018; Colarco et al., 2003; Husar et al., 1997; Kandler et al., 2018; Zuidema et al., 2019). Although to the south of this study's domain of the WNAO, the Puerto Rico Dust Experiment (PRIDE) is especially noteworthy as it represented a very thorough campaign to characterize mineral aerosol properties and cycling over the WNAO (Colarco et al., 2003; Levy et al., 2003; Reid et al., 2003a; Reid et al., 2003b). There are many unknowns associated with the characteristics of dust arriving to the WNAO and their ultimate impacts. Past work showed that a global chemical and transport model (GEOS-Chem) underestimates transport of dust to the Western Atlantic and/or has too much deposition over the Caribbean Sea (Generoso et al., 2008). An intercomparison study for several global models participating in the Aerosol Comparison between Observations and Models (AeroCom) experiments revealed significant discrepancies in dust amount and distribution over a broad region containing the North Atlantic Ocean, urging for the need for more observational data to constrain model processes (Kim et al., 2014 and references therein). An open area of research is how dust properties change upon their transport from source regions such as the Saharan Desert along their trajectory to the WNAO with intermediate stops at research-active regions such as Barbados. More specifically, efforts are needed to characterize how the chemical, optical, and hygroscopic properties of dust change via potential cloud and clear-air processing and after continued long-range transport. These processing mechanisms are critical as well with regard to potentially converting insoluble forms of ocean micronutrients such as Fe into more soluble forms.

The vertical distribution of dust over the WNAO is also a critical gap (Colarco et al., 2003; Peyridieu et al., 2010) that is relevant to a number of issues such as impacts on clouds, vertical thermodynamic profiles, and removal processes. A recent modeling study that examined transport pathways of dust to the Northeast U.S. showed that the majority of plumes reside between 2 to 6 km, but urged that more observational data are needed to examine vertical dust structure (Zhang et al., 2019). Dust is thought to have the potential to impact the North Atlantic Ocean circulation, specifically large-scale barotropic and overturning circulations (Serra et al., 2014). These large-scale phenomenon that may have sensitivity to dust certainly require more investigation owing to their importance for the region's climate system and ocean processes.

One additional fundamental research need for the WNAO is to characterize all sources contributing dust to the region and the relative importance of these sources as a function of season. While most of the attention thus far has been on North African dust, there likely can be contributions from other sources such as North America and transported biomass burning plumes that have been shown to be carriers of soil dust (Maudlin et al., 2015; Schlosser et

al., 2017). Additional impacts and questions surrounding biomass burning aerosols are presented in the next section.

5.2.2 Biomass Burning—A number of studies have characterized the spatial distributions and physicochemical properties of biomass burning plumes over the Eastern U.S. and North Atlantic Ocean, especially in field campaigns already mentioned such as ICARTT (Clarke et al., 2007; Cook et al., 2007; de Gouw et al., 2006; Heald et al., 2006; Lewis et al., 2007; Martin et al., 2006; Peltier et al., 2007; Sullivan et al., 2006; Thornhill et al., 2008) and TCAP (Muller et al., 2014). Sources of such plumes, thus far mostly documented for summer months, are from distant upwind regions such as Alaska, Western and Central Canada, Western U.S., and even Siberia (Honrath et al., 2004; Li et al., 2005; Owen et al., 2006; Parrington et al., 2012; Yang et al., 2018). Plumes transported from those regions have the ability to significantly impact surface sites along the North America East Coast (Millet et al., 2006; Rogers et al., 2019). The WINTER campaign suggested that most biomass burning signatures in the study region during the winter months likely stem from regional sources in the form of residential wood burning (Schroder et al., 2018; Sullivan et al., 2019a). Investigations associated with biomass burning are becoming increasingly important owing to increased prevalence of fires associated with warming and drying (Gillett et al., 2004; Kasischke & Turetsky, 2006), in addition to human impacts (Mollicone et al., 2006).

While it has been well-documented that emission injection heights of fires require better characterization (Pfister et al., 2006), characterizing their vertical profiles over the WNAO is of great importance with regard to vertical heating rates and stability (Taubman et al., 2004) and interactions with clouds. Studies in other regions such as the southeastern Atlantic (Rajapakshe et al., 2017) and northeastern Pacific Ocean (Mardi et al., 2018) have looked at vertical characteristics such as the distance between plumes and cloud top heights. Similar work is warranted for the WNAO in pursuit of better understanding of regional ACI and subsequent effects on radiative forcing.

5.3 Atmospheric Deposition

The majority of deposition studies reviewed for the study region have focused on wet deposition. The North American East Coast has had the benefit of long-term monitoring via networks such as the Canadian Air and Precipitation Monitoring Network (CAPMoN) and the National Atmospheric Deposition Program/National Trends Network (NADP/NTN). Furthermore, many studies have focused on measurements at Bermuda. Long-term research at this remote island has provided an understanding of atmospheric chemistry processes, transport processes, and speciated concentrations (Altieri et al., 2016; Altieri et al., 2013; Church et al., 1984; Galloway et al., 1993; Jickells et al., 1998; Keene et al., 2015; Knapp et al., 2010; Lim et al., 1994; Moody & Galloway, 1988; Sholkovitz et al., 2010; Tian et al., 2008). One prime example of the importance of measurements from Bermuda is shown in the work of Keene et al. (1986) who had perhaps the first published report of the uncertainties associated with calculating sea salt and non sea salt components for wet deposition samples, which arise from factors such as variability in seawater composition, assuming purely marine sources for sea salt reference species, and analytical uncertainties.

Data from Bermuda and other remote marine areas provide a benchmark of precipitation composition and wet deposition fluxes that can be compared with coastal regions dominated by anthropogenic processes, such as the populated and industrialized regions along the East Coast of North America. In contrast to continental measurements of wet deposition over the East Coast and Bermuda, shipboard measurements (on windward side) have provided the advantage of contamination-free data (Galloway et al., 1983) and spatial information over the ocean where surface sites are lacking (Berresheim et al., 1991; Church et al., 1991). Wet deposition long-term trends have shown higher levels of SO_4^{2-} , NO_3^- , and trace metals (e.g. Pb, copper (Cu), and Fe) in populated areas over North America when compared to remote areas over the WNAO (Church et al., 1984; Galloway et al., 1983; Galloway et al., 1982; Jickells et al., 1982; Keene et al., 2015; Veron et al., 1992). However, similar to the aerosols measured at Bermuda, a large fraction of the nss SO_4^{2-} measured in precipitation at Bermuda is primarily due to transport from the North American East Coast (Keene et al., 2015).

An advantage of routinely collected data at stations associated with the CAPMoN and NADP/NTN along the U.S. and Canada coasts has been analysis of long-term trends such as reductions in SO_4^{2-} (Driscoll et al., 2001). Although changes in precursor emissions have been documented as the root cause of SO_4^{2-} reductions, Dayan and Lamb (2008) showed that temporal changes in large-scale circulation patterns should be considered as well in driving trends in summertime concentrations along the East Coast. As SO_4^{2-} concentrations have decreased over the U.S. due to pollution regulations, a similar trend was observed in Bermuda based on data from 1989 to 1997 and from 2006 to 2009 (Keene et al., 2014). Two more interesting results published from this long-term dataset were that: (i) unlike nss SO_4^{2-} , precipitation concentrations of NO_3^- at Bermuda have not shown a similar decrease associated with regulatory actions to reduce NO_x emissions, which is thought to be due to efficient processing of oxidized N in marine air, and (ii) NH_4^+ concentrations in precipitation increased significantly over recent decades owing possibly to increasing NH_3 emissions from adjacent continental regions. It is important to continue monitoring these ion concentrations as a decade has passed since the final year of the data record used by Keene et al. (2014). Increased measurement capability for organics and black carbon in wet deposition samples can help provide a broader view of the chemical profile of precipitation (Barrett et al., 2019). In this regard, continued efforts to quantify organic N in wet deposition is warranted as was done by others over parts of the U.S East Coast showing appreciable contributions to total dissolved N (Keene et al., 2002b; Scudlark et al., 1998). Case study events should also be a point of future research with wet deposition data, which has been helpful in the case of learning about precipitation chemistry during hurricanes, especially for the southeastern U.S coast (Mullaugh et al., 2013) and along the northeastern U.S. coast (Benitez-Nelson & Buesseler, 1999; Raymond, 2005).

A significant and persistent knowledge gap in our understanding of geochemical cycles and the life cycle of atmospheric constituents involves dry deposition. Dry deposition of gases and aerosols onto land or water bodies contributes to water quality, both terrestrial and marine physicochemical and biogeochemical processes, and marine productivity. The FAMS investigation was a coordinated effort over Florida to examine both dry and wet deposition of Hg, but also other species such as sea salt, dust, urban pollution, and acidic aerosols

(Landing et al., 1995). Pai et al. (1997) reported based on model simulations that the wet deposition of Hg was twice that of dry deposition. The dry deposition of N-containing pollution has been of special interest for the Chesapeake Bay estuary and WNAO coastline as it is a major source of N (Loughner et al., 2016). Because coastal city pollution flows offshore to accumulate over coastal waters where there is an extra input from shipping emissions, followed by onshore flow again, there can be enhanced dry deposition of N assisted in part by faster deposition over land than water (Loughner et al., 2016). Sources of this pollution include ammonium nitrate from agriculture and sodium nitrate formation over waters with sea salt emissions (Loughner et al., 2016). Dry deposition accounted for approximately 43% of total atmospheric N deposition based on a 2-week sampling period at Lewes, Delaware on the mid-Atlantic coast (Russell et al., 2003). In that study, NH₃ was shown to account for the majority of the total dry deposition of N (~60%). Several studies at Appledore Island, Maine during ICARTT provided important quantitative results associated with dry deposition of HNO₃ (Fischer et al., 2006), inorganic chlorine and bromine (Keene et al., 2007b), and NH₃ (Smith et al., 2007).

Although deposition of dust has been examined in areas such as Florida (Muhs et al., 2007) and Bermuda (Muhs et al., 2012), models treating dust deposition have not been able to be validated extensively owing to a scarcity of long-term measurements of dust deposition rates (Prospero & Landing, 2009). Prospero et al. (2010) showed that models quantifying the ratio of wet deposition to dry deposition of dust had widely ranging ratios (1:1 to 30:1), in sharp contrast to FAMS data (3:1 to 4:1).

Over the open ocean, interest about dry deposition of species such as N is significant partly because these inputs have increased dramatically over the last several decades. For instance, Baker et al. (2010) noted that N inputs to the open ocean have increased by up to a factor of three over the last 150 years with more enhancements expected in the near future. But their calculations, as with others such as the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Baker et al., 2017), are difficult to validate since lack of data results in dry deposition calculations over the ocean to be based largely on emissions and transport modeling. Furthermore, there are major uncertainties in dry deposition velocities, which are needed to quantify deposition fluxes from concentrations (Baker et al., 2017). This latter issue applies broadly to all atmospheric constituents that can deposit to the surface, including of course sea salt aerosol as was demonstrated during EOPACE (Reid et al., 2001).

Although not unique to the WNAO and East Coast, a better understanding of deposition velocities will benefit estimations of deposition fluxes over the ocean. Over land, monitoring networks such as the EPA Clean Air Status and Trends Network (CASTNet) and the NOAA Atmospheric Integrated Research Monitoring Network (AIRMoN) have shown value in examining dry deposition (Driscoll et al., 2001). However, more long-term monitoring efforts, including gas and aerosol speciation and deposition fluxes, are needed since cruises over open waters have provided measurements for only short periods of time. For aerosols in particular, size-resolved concentrations are arguably most needed.

5.4 Clouds and Storms

As noted earlier in Section 2, ETCs are of importance to the U.S. East Coast. A substantial body of work has examined aspects of East Coast winter storms, which have major societal impacts ranging from commerce to public safety over the mid-Atlantic and northeastern U.S. (e.g., Brennan & Lackmann, 2005; Charles & Colle, 2009a, 2009b; Evans & Jurewicz, 2009; Ganetis & Colle, 2015; Hirsch et al., 2001; Kocin & Uccellini, 2004; Kumjian & Lombardo, 2017; Nicosia & Grumm, 1999; Novak et al., 2004; Novak & Colle, 2012; Root et al., 2007; Sanders & Bosart, 1985; Zhang et al., 2002). In fact, the oldest study identified in this review paper is related to cyclogenesis over the East Coast, which is relevant to the modeling and forecasting of such storms (Miller, 1946). Various studies have examined individual events with considerable impacts on parts of northeastern U.S., including the 9 February 2013 blizzard over the Northeast (Picca et al., 2014), and various snowstorms in winter months, ranging from October to April, of other years over the last few decades (Hirsch et al., 2001; Nicosia & Grumm, 1999).

As summarized by Greybush et al. (2017), practical predictability of these storms is limited based on the collective use of observations, models, and assimilation systems with a significant barrier being the mesoscale characteristics of snowfall patterns that frequently take the form of banded structures (Greybush et al., 2017; Molthan et al., 2016; Novak et al., 2004; Novak et al., 2010). Mesoscale precipitation bands often are observed in what is commonly referred to as the “comma-head” portion of ETCs in the Northeast (Nicosia & Grumm, 1999; Novak et al., 2004; Novak et al., 2010). To augment decades of research trying to characterize the nature of these storms and their associated precipitation, the upcoming Investigation of Microphysics and Precipitation for Atlantic Coast-Threatening Snowstorms (IMPACTS) mission will study winter snowstorms over the Northeast U.S. and offshore areas. IMPACTS will provide observations critical for the understanding of the formation and evolution of snowfall and associated banded structures, and will apply the observations to improve snowfall remote sensing and modeling.

A significant amount of cloud research over the WNAO has also been devoted to CAOs, which are an important feature during the winter season, with an example shown from 22 January 2019 in Figure 1. The Gulf Stream domain is identified as one of the regions that is most likely to have very strong CAOs based on a global analysis (Fletcher et al., 2016a). As discussed in Section 3, an extensive number of studies examined characteristic features during CAOs several decades ago (Atlas et al., 1983; Bosart, 1981; Bunker, 1976; Bunker & Worthington, 1976; Chou & Atlas, 1982; Gall & Johnson, 1971; Ichiye & Zipser, 1967; Sethuraman et al., 1986; Sweet et al., 1981). These original studies laid the framework for major campaigns in later years such as GALE in 1986. CAOs have been examined on a case study basis over the WNAO by many using various types of datasets (e.g., satellite, rawinsonde, mooring, buoy) (Babin et al., 2003).

As WNAO research has shown, CAOs occur when cold, stable, and stratified air originating over land or ice is transported over the warmer ocean surface, leading to enhanced surface heat fluxes over synoptic-scale areas (Raasch, 1990). A convective boundary layer then develops leading to horizontal roll vortices (e.g., roll streets in a helical roll circulation) with the roll being aligned in the direction of the mean low-level wind (Young & Sikora, 2003). A

zone eventually develops with closed- or open-cell clouds. An opportunity provided by CAO events, as demonstrated by recent studies over the Eastern Atlantic Ocean (e.g., Wood et al., 2017), is to study features associated with ultra-clean aerosol conditions (Field et al., 2014). Such conditions are desirable since, similar to the current day Southern Ocean, they mimic the atmospheric state reminiscent of pre-industrial times that serve as the basis for critically important effective radiative forcing calculations (Hamilton et al., 2014).

Important issues that remain are linked to the ability of GCMs to adequately simulate cloud cover in Northern Hemisphere CAOs and also the post cold frontal (PCF) zone of Southern Hemisphere (SH) midlatitude cyclones (Bodas-Salcedo et al., 2014; Field et al., 2014; Williams et al., 2013). The failure to simulate clouds in SH midlatitude cyclones creates a significant bias in the absorbed shortwave radiation (Trenberth & Fasullo, 2010) affecting the entire SH energy budget (Hwang & Frierson, 2013). It is for this reason that there has been a particular interest in using field experiments on relatively well observed NH CAOs for climate model evaluation and development (Bodas-Salcedo et al., 2012), while similarities and differences between NH and SH CAOs have been well-documented (Fletcher et al., 2016a; Fletcher et al., 2016b). The role of cloud microphysics (and by extension, aerosols) in affecting the morphology, vertical, and horizontal extent and thermodynamics of the cloud fields in CAOs remains unclear (Abel et al., 2017; Field et al., 2014). However, one recent study identified the spatial extent of a CAO as being a primary control on cloud cover, while the strength of turbulent surface fluxes determined the liquid water path and cloud vertical extent (Naud et al., 2018). In addition, CAO cloudiness is observed to exhibit downwind transitions from stratiform (generally near the coast) to cumulus once the MBL has deepened. Questions remain surrounding the drivers for these transitions, their representation in models, and their impact on cloud radiative effects (Naud et al., 2019).

CAO events also provide a useful meteorological constraint (e.g., widespread and sometimes quasi-stationary cold air advection, dry and relatively cloud-free mid- and upper-troposphere and persistent offshore surface flow from the northwesterly quadrant) over the WNAO that can facilitate investigation of ACI without the confounding impact of more complex transport patterns. Of particular interest is the opportunity that CAO conditions provide to investigate the observed gradient in anthropogenic pollution proxies with offshore distance. In addition, recent work shows that the largest fine mode AODs associated with ETCs are in the PCF zone and that variations in cloud cover that are correlated with aerosol variations in the PCF zone show less dependence on cyclone strength and humidity than in other areas of cyclones (Naud et al., 2016, 2017).

Of note with regard to CAOs is the upcoming Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE), which will be discussed in more detail in the following sections, that aims to improve understanding of the continuum of warm MBL clouds ranging from stratiform to cumulus in the WNAO, with a subset of flights planned to specifically target CAOs (Sorooshian et al., 2019).

5.5 Air-Sea Interactions

Some field experiments devoted to understanding air-sea interactions over the WNAO, as reviewed in Sections 3–4, have provided measurements of air-sea fluxes (or covariance estimates) and/or primary state variables and parameters for various flux terms (see background on bulk formulae in Section S1 of Supplement) near or away from strong boundary currents (e.g., the Gulf Stream). However, the spatial scales of variability in MBL meteorological variables, on which the processes that link the meteorology and clouds are operating, are critical but poorly understood over the WNAO. Using buoy array data from FASINEX, Weller et al. (1995) found no gradients in winds or clouds that could be ascribed to localized circulation or convection associated with mesoscale variability in SST and air-sea fluxes, which was inconsistent with results from previous studies and pre-campaign modeling results.

The western boundary current of the North Atlantic advects warm waters along the entire U.S. East Coast, with temperature differences of 5 – 10 K occurring across only a few km at times (Warnecke et al., 1971). This feature can alter the cloud field through other processes in addition to CAOs. The SST gradients near the Gulf Stream, resulting in strong heat flux gradients, can give rise to circulations resembling sea-breeze fronts (Sublette & Young, 1996), with higher cloud tops towards the warm side of the front (Liu et al., 2014). The warmer SSTs can reduce the local atmospheric stability, affecting the low cloud field and modulating the surface winds through the vertical momentum flux (Chelton et al., 2004). The gradients in heat flux are important in modulating synoptic atmospheric variability and meteorological states of the boundary layer. For example, following the passage of a CAO, westerly to northerly flow of cold and dry continental air moving over the warm Gulf Stream can generate a total sensible and latent heat flux in excess of 1000 W m^{-2} from the ocean to the atmosphere (Bane & Osgood, 1989). In response to such large heat loss in the upper ocean layer, the Gulf Stream was found to have a cooled and deepened mixed layer. Seasonal responses to air-sea interactions involving the Gulf Stream can also be observed over the WNAO. A springtime phenomenon features very long cloud lines (~1000 km) forming over the major axis of the Gulf Stream. Previous modeling studies ascribe these lines to the large surface thermal gradient (Li et al., 2004). Another phenomenon giving rise to low cloud formation during the wintertime has been called the ‘Gulf Stream Front’, wherein continental cold air underlying warmer, moister air offshore quickly saturates, developing stratus or ‘sea fog’ (Carson, 1950).

Air-sea interactions can also involve the exchange of gas species, sea spray, and other forms of marine-derived particles at the wavy interface of the ocean. The ocean contains substances like C, N, O, and P which impact Earth climate through biogeochemical processes. Due to the warming effect of rising concentrations of atmospheric CO_2 , questions regarding the carbon cycle have received increasing scientific attention over recent decades. The ocean exchanges CO_2 with the atmosphere and acts along with the terrestrial ecosystems as the most important CO_2 sink. As discussed in Section 5.2.1, atmospheric dust from North Africa that deposits in the WNAO is a major supplier of Fe and P nutrients to open ocean and terrestrial ecosystems. DMS and primary marine aerosols emitted from the

WNAO surface can significantly affect atmospheric aerosol concentrations, cloud droplet/ice nucleation, surface precipitation, and the energy budget.

Of paramount importance is the flux of primary marine aerosols, which was discussed in Section 5.2. Production fluxes of primary marine aerosols are an area still requiring research in terms of fully comprehending the key governing factors and their relative importance and interactions. Wind speed alone is insufficient to model sea spray fluxes (e.g., Reid et al., 2001), while it has been shown that other controlling variables can include wave kinematic characteristics including air entrainment with breaking waves (Long et al., 2011), SST (Jaegle et al., 2011; Lewis & Schwartz, 2004), and ocean salinity (Sofiev et al., 2011). A motivator for advancing understanding of primary marine aerosol production fluxes, and associated source function parameterizations, is that these generated particles impact a host of important reactive constituents such as halogens and organics, as well as impacting a number of processes linked to radiative transfer, cloud formation, precipitation, and biogeochemical cycles. Further characterization of the constituents enriched in these generated aerosols by bursting bubbles is important to continue learning about the enhancement of organics as was shown convincingly in several studies linked to the R/V Endeavor cruise between September and October 2016 over the WNAO (Beaupré et al., 2019; Frossard et al., 2019a; Frossard et al., 2019b).

There are several remaining challenges in quantifying air-sea fluxes and understanding their impact on MABL clouds over WNAO. Aside from topics associated with deposition covered in Section 5.3, selected areas warranting future research include: (i) Development of scale-dependent surface turbulent flux parameterizations that work for various model resolutions (from LES to climate models), (ii) Consideration of convective gustiness and fluxes under extreme wind conditions, (iii) Resolving air-sea fluxes near the Gulf Stream (strong SST gradients), (iv) Resolution of issues surrounding coarse resolution satellite retrievals. (For example, the 25 – 50 km spatial footprint of microwave satellite SST retrievals is too coarse to resolve the gradients near the Gulf Stream and potentially suffers land contamination in coastal regions.), (v) Advancing the ability to quantify near-surface air temperature and humidity with satellite sounders, (vi) Development of methodology for consideration of sea spray droplet evaporation effects on latent heat and moisture fluxes, which is at the moment still poorly understood and difficult to consider in estimates (Fairall et al., 2003), (vii) Estimating ocean surface currents, which are strong over the western boundary currents region, challenging to measure and represent in numerical models, and are needed to calculate wind speed and wind shear, (viii) Resolving issues arising from temporal offsets in measurements of surface fluxes, state variables, and clouds for the study of the impact of air-sea interactions on MBL clouds, and (ix) Improving understanding of size- and composition-dependent production fluxes of sea spray aerosol (including organics and sea salt) and predicted distributions of MBL CCN (de Leeuw et al., 2011).

5.6 Development/Validation Efforts

Studies over the extended WNAO region have taken advantage of its unique geographic location in order to test models of varying complexity, to improve remote sensing retrievals, and to demonstrate new techniques and instrumentation. Section 4.5, for instance, provided a

snapshot of these types of activities over Bermuda, while many more efforts of this type are summarized in Tables 1 and S1. Several avenues of future development and validation can be of great help for WNAO research. Here we focus on recent examples related to remote sensing developments.

During TCAP, coincident measurements of aerosol properties were conducted by two aircraft, specifically the DOE Gulfstream-1 and the NASA LaRC B-200 King Air. The combination of the NASA High Spectral Resolution Lidar-2 (HSRL-2), which can measure particle backscatter coefficients at 355, 532, and 1064 nm, and the Gulfstream-1 aerosol payload were used to evaluate multiwavelength lidar retrievals of aerosol properties, such as particle size, through a novel inverse retrieval algorithm (Muller et al., 2014). This type of effort can be enhanced even further by incorporating the effects of hygroscopic growth as well as refractive index determination (Sawamura et al., 2017). Refractive indices have been shown to be undercharacterized over the WNAO and other regions in general (e.g., Aldhaif et al., 2018; Ferrare et al., 1998; Shingler et al., 2016). These novel techniques represent an advance upon the prior methods that related column-integrated AOD to aerosol and CCN concentrations (Andreae, 2009; Shinozuka et al., 2015) or passive remote sensing techniques requiring strong assumptions as to cloud properties (Rosenfeld et al., 2016). Most of the past work in this area has focused on continental regions with relatively high aerosol loadings; however, an important scientific need is to extend and evaluate these techniques to remote marine and coastal mixed-pollution environments (i.e., WNAO) where there are fewer in situ observational constraints on the total aerosol and CCN budgets.

Another advancement that is ideal for the WNAO region is the combined deployment of a lidar and polarimeter on an airborne platform, which is a top priority for advancing aerosol-cloud science according to the National Academies of Sciences, Engineering, and Medicine (2018). This type of effort has taken place before in recent campaigns in the region such as NAAMES and TCAP (Behrenfeld et al., 2019; Berg et al., 2016). Together, these remote sensors can advance the ability to retrieve cloud-top values of N_d and liquid water content without assumptions about cloud adiabaticity, in addition to being able to quantify autoconversion rates and cloud-top mixing processes. The upcoming ACTIVATE mission over the WNAO will jointly deploy a HSRL-2 and a RSP on a King Air platform.

5.7 Aerosol-Cloud Interactions

Although ACI have been pinpointed by recent reports (IPCC, 2013; National Academies of Sciences & Medicine, 2018) to be associated with the largest uncertainty in estimating global anthropogenic radiative forcing, there have been few studies over the WNAO to examine such interactions in detail. More specifically, Table 1 shows that out of the ~710 published works identified for the WNAO region, only 24 by our count directly examined ACI topics. This is in sharp contrast with the western coast of the U.S. where the majority of past research studies have investigated ACI (Sorooshian et al., 2018 and references therein), many of which were rooted in understanding the nature of ship tracks (Russell et al., 2013 and references therein).

ACI studies over the WNAO can be categorized into those that studied “cloud effects on aerosols” and “aerosol effects on clouds”. A fair number examined the former issue owing to

the great interest in wet scavenging over the extended WNAO region (e.g., Altieri et al., 2016; Galloway et al., 1993). Todd et al. (2003) showed that precipitation removes optically active aerosols at Bermuda, which in turn alters spatiotemporal variability in the Earth's radiative balance. de Gouw et al. (2006) showed that a precipitation event led to a reduction in particle volume concentration and also acetic acid, which is a soluble VOC. Garrett et al. (2006) used airborne data to demonstrate that ratios of soluble and insoluble trace gases can yield estimates of scavenging of CCN by precipitating clouds. Weigelt et al. (2009) probed how cloud contact time impacted aerosol size distribution characteristics, with different responses observed in the northern and southern parts of the WNAO.

With regard to past WNAO studies of how aerosols affect clouds, two airborne investigations as part of NARE (Leaitch et al., 1996) and SOLAS (Leaitch et al., 2010) specifically focused on N_d and controlling factors such as degree of turbulence and abundance of carbonaceous aerosol. Medina et al. (2007) conducted a surface-based CCN closure study at the Thompson Farm AIRMAP site in New Hampshire and concluded that size-resolved chemical data and treatment of mixing state are helpful for more successful closure. Satellite-based studies have shown that the strength of the aerosol indirect effect (AIE; $-\ln(r_c) / \ln(\tau_a)$, where r_c = droplet effective radius, τ_a = AOD, and evaluated in conditions of fixed cloud liquid water) exhibits seasonal dependence and is highest in summer due to the influence of continental outflow (Bulgin et al., 2008; Jones et al., 2009). Jones et al. (2009) further showed that wintertime is associated with the highest cloud fractions and cloud thicknesses, and that AIE was higher in the WNAO for thicker clouds. Avey et al. (2007) used satellite data and a trajectory model to conclude that the expected AIE responses (smaller r_c with increased aerosols at fixed liquid water) were observed for a period during the ICARTT campaign (summer 2004) for North American continental outflow close to the coast but that such a response became absent within 4 ± 1 days of advection due to likely wet scavenging of CCN. They observed no change in cloud liquid water path due to enhanced anthropogenic aerosols. MODIS data analysis in June-August of 2002 showed that increased levels of aerosols between 30°N and 60°N over the Atlantic Ocean were linked to an increase in shallow cloud coverage (Kaufman et al., 2005). Liu and Li (2019) suggested that the dependence of cloud development on lower tropospheric stability (LTS) is weakened as a function of aerosol loading based on a year of TCAP data over Cape Cod, Massachusetts.

A past study of weekly cycles over the WNAO found intriguing trends for air pollutants, rain, and tropical cyclones (Cerveny & Balling, 1998). They suggested that the following observed weekly cycles were driven by the thermal influence of continental aerosols: (i) less pollution early in the week, (ii) more rain on weekends at near-coastal areas, and (iii) near-coastal cyclones exhibit weaker surface winds and higher frequency on weekends. Booth et al. (2012) showed that aerosols, especially during periods of volcanic activity, explain the majority (76%) of multidecadal variance in detrended 1860 – 2005 SSTs in the North Atlantic and that ACI dominated the spatial pattern and magnitude (80%) of total surface aerosol forcing and SST spatiotemporal variability. While substantial attention has been given to the interactions between aerosols and hurricanes in the Eastern North Atlantic region, especially with regards to Saharan dust impacts (Chiacchio et al., 2017; Fontenot, El-Askary, Garay, Campbell, & Kalashnikova, 2018; Nowottnick et al., 2018; Sauter &

L'Ecuyer, 2017; Schwendike, Jones, Vogel, & Vogel, 2016), there has been very limited attention given to these relationships in the WNAO.

The transition between the continentally-influenced coastal atmosphere to the offshore WNAO marine atmosphere results in a longitudinal gradient in aerosol concentrations (Figure 5), composition, and physical properties, and hence the ability of these aerosols to act as CCN to form cloud droplets. Modeling studies suggest that the aerosols would become more hygroscopic as they move farther offshore with a mean hygroscopicity parameter (κ) value of ~ 0.3 over North America and ~ 0.59 over the North Atlantic Ocean (Pringle et al., 2010). Against the backdrop of decreasing overall aerosol concentrations, the impact of these compositional shifts for the CCN budget and droplet activation is unclear. Recent modeling work examining the sensitivity of N_d to changes in aerosol concentrations ($\ln N_d / \ln N_a$) indicates a shift in from near 0 over land (so-called 'aerosol rich' conditions) to close to 0.5 over the WNAO, where aerosol loadings are much lower (Moore et al., 2013). Building off that study, Figure 6 shows a spatial map specifically for the WNAO, which shows the spatial distribution of N_d and cloud albedo sensitivity relative to the strong aerosol gradient. Both sensitivities become enhanced farther offshore, especially east of Bermuda. While many aerosol-cloud observations have been made over relatively polluted continental regions, the observational data set for transitional aerosol regimes such as the broad WNAO region remain limited to date. There is a need for additional observations to evaluate the skill of such modeling predictions.

There have been extensive ACI-relevant measurements conducted in regions adjacent to the WNAO, such as with NAAMES (Behrenfeld et al., 2019) and ACE-ENA (Wang et al., 2019a) in the Northern and Eastern North Atlantic Ocean regions, respectively. Timing is appropriate to accelerate efforts for the WNAO to build more knowledge about ACI, especially since this region is more meteorologically complex than the traditional stratocumulus regions investigated that offer the limited view of a single cloud regime. A new avenue of inquiry could be learning if high background aerosol concentrations and/or the complexity in meteorology off the U.S. East Coast are the reason for why ship tracks visible from space are not a frequent feature in contrast to the U.S. West Coast. Previous ACI process modeling studies (Possner et al., 2018; Wang et al., 2011) have shown that the formation of visible ship tracks depends on background aerosol concentrations as well as cloud regimes determined by meteorological conditions such as MBL structure, humidity, wind shear, and surface turbulent fluxes. Furthermore, aerosol loadings have diminished in recent decades over the WNAO, as noted above, which inevitably can pose a change in ACI for the region (Jongeward et al., 2016). Examining how the nature of these interactions has changed over time can yield important insights about the impacts of regulatory activities beyond how total concentrations of species like SO_4^{2-} and NO_3^- have been reduced.

ACI processes are highly complex (Stevens & Feingold, 2009) and model representations are highly varied, yielding large uncertainties in estimates of future impacts on weather and climate. Process models can find contradictory outcomes depending on the processes and environmental regimes that are emphasized. Many studies show competing effects of aerosol on cloud water, which is the largest lever on cloud radiative effects. For example, Albrecht (1989) proposed that a reduction in cloud drop size from increased aerosol concentration

(for a constant liquid water) would increase cloud lifetime and water path through a suppression of precipitation; on the other hand, Ackerman et al. (2004) showed that a dry layer above cloud could result in a decrease of cloud liquid water and optical thickness through entrainment and evaporation. To accurately diagnose aerosol effects on cloud processes, a comprehensive characterization of the cloud, aerosol, and dynamical conditions is required, and sometimes at both the cloud and mesoscale. A key outcome of ACTIVATE will be a detailed, statistical population of ACI-relevant measurements gathered over several years of deployment with a comprehensive suite of measurements from two aircraft.

6. Conclusions

Here we have provided a comprehensive review of atmospheric research over the WNAO and North American East Coast since the 1940s to the present. There have been over 50 different field projects and long-term monitoring efforts, in addition to 715 peer-reviewed publications between 1946 and 2019. The timeline of research over the WNAO, North American East Coast, and Bermuda were summarized, in addition to a lengthy list of selected results and areas requiring more research based on findings of past works. Here a final set of recommendations is provided that is not exhaustive but isolates a few selected issues from the longer list of recommended efforts in Section 5. It is proposed here that a higher priority should be directed towards interdisciplinary research topics, followed by disciplinary topics, and lastly continued efforts in development and validation projects.

• Interdisciplinary Research

The areas with the most potential for advancement require synergistic collaboration between investigators specializing in numerous disciplinary topics. Two examples of topics involving high complexity are provided here that are considered to be of relatively high importance for the WNAO region. The first example relates to continued research in air-sea interactions, which involves knowledge of numerous processes including (but not limited to) surface heat and moisture fluxes, primary marine aerosol fluxes, atmospheric deposition, and ocean productivity. Research to tie together these pieces and their ultimate impact on radiation, boundary layer processes, clouds, and precipitation is needed owing to their interconnected nature. The coupling and co-location of marine aerosol production and turbulent fluxes is important for the second interdisciplinary topic, ACI.

ACI is the topic examined in this review paper with the least amount of publications (24 out of 715). Furthering understanding of ACI over the WNAO requires systematic and robust measurements of meteorological/state variables, aerosols, and cloud properties in combination with numerical modeling of air-sea interactions and cloud/fog formation processes (e.g., Sorooshian et al., 2019). In addition to such observations, the metrics used to quantify ACI must simultaneously evolve to embrace the complexity of the aerosol-cloud process system. Until now, metrics that build understanding of aerosol impacts on cloud properties take a ‘chain approach’ (Ghan et al., 2016), which captures the primary relationships between aerosol and the cloud drop activation process very well. However, there is limitless interconnectedness among cloud-scale dynamical processes (e.g., updraft velocity, entrainment), aerosol processes (e.g., composition-dependent hygroscopicity,

radiative heating), and larger-scale meteorological processes (e.g., moisture advection, surface-atmosphere fluxes) that combine to produce cloud radiative and precipitation properties in any given situation. As just one step forward of many steps needed, the upcoming ACTIVATE project is accommodating this extensive complexity by developing new methods using various approaches such as process-based metrics (e.g., Jing & Suzuki, 2018) to constrain model representations and multi-variate approaches to characterize the larger system.

• **Disciplinary Research:**

Examples of priorities for disciplinary research include studies along the lines of Sections 5.1–5.3 where various aspects of atmospheric chemistry and microphysics are considered. As motivated already, the reduction in certain constituents over North America due to regulatory activities, coupled to the ubiquity of sea salt over the WNAO highlights the importance of halogen radical chemistry. As highlighted in Section 5.1, there are important opportunities laying ahead for the WNAO region to examine the nature of halogen radical cycling and its overall impact on tropospheric chemistry, radiation, clouds, and precipitation. The sensitivity of these halogen species to large-scale circulations and their transfer between the MBL and free troposphere requires more investigation.

It is challenging to constrain the MBL CCN budget in the WNAO region owing to difficulty in deconvoluting seasonal variability in local meteorology (MBL structure, thermodynamics), variability in emissions sources (pollution transport, surface fluxes, free tropospheric entrainment), and variability in aerosol loss mechanisms (precipitation scavenging, air mass history, mesoscale/synoptic scale meteorology). It is thought that the CCN budget in the remote MBL is governed largely by emissions of organics and sea salt in sea spray in addition to subsidence of free tropospheric air that can contain DMS-derived species and other continentally-derived aerosol constituents (Quinn & Bates, 2011). In agreement with Quinn et al. (2017), an important knowledge gap to fill is identifying the relative importance of nss SO_4^{2-} sources impacting the MBL (e.g., DMS oxidation in the MBL, entrainment of free tropospheric air containing oxidized DMS, transport of anthropogenic emissions in either the MBL or free troposphere). One way to try to address this is with long-term measurements of size and composition with the ability to differentiate between biogenic and anthropogenic SO_4^{2-} covering different seasons and regions.

Continued focus on primary marine aerosol fluxes is needed to extend the strong foundation of work by several investigators over the region that has raised new questions into factors such as surfactant properties (Frossard et al., 2019a), characterization of physicochemical properties and fluxes of primary marine aerosol over the full spectrum of aerosol sizes (Keene et al., 2017), and further consideration of the various factors affecting fluxes (e.g., biogeochemical and physical processes in sea water, time of day and season, characteristics at the air-water interface) to improve source functions. The nature and role of organics enriched in marine aerosols also warrants further examination owing to subsequent impacts on radiative and hygroscopic properties of the aerosol, and also general multiphase photochemical processing in the MBL (Zhou et al., 2008).

• Development/validation efforts

Technological advancements, improved techniques for data analysis, and model intercomparison and development efforts with observational products are all necessary to advance the ability to examine the atmospheric complexity of the WNAO region and North American East Coast. Another type of advancement in terms of how to approach science topics for the region is in the design of airborne campaigns to collect more statistics, gather vertically-resolved information, and combine in situ and remote sensing methods. This approach is embraced by the upcoming IMPACTS and ACTIVATE (Sorooshian et al., 2019) missions introduced in Section 5.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Key Points:

1. 50+ field studies and 700+ papers illustrate the complexity of atmospheric phenomena over the W. North Atlantic and N. American East Coast
2. The widest body of work has been devoted to atmospheric chemistry and has characterized urban outflow and marine emissions
3. Multi-disciplinary topics such as aerosol-cloud and air-sea interactions have not been sufficiently addressed and warrant high priority

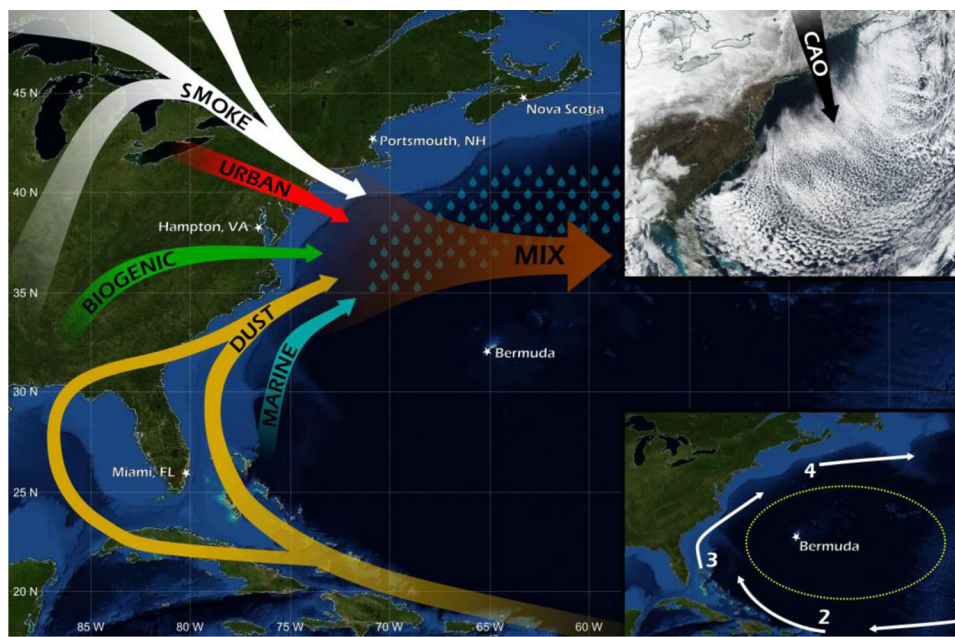


Figure 1. Map of the extended Western North Atlantic Ocean (WNAO) with major features shown such as key sources of gases and aerosols, an example of a cold air outbreak (CAO), and ocean currents that transport water around the Atlantic Ocean to form the North Atlantic gyre, within which lies Bermuda and the Sargasso Sea (1 = North Equatorial Current, 2 = Antilles Current, 3 = Gulf Stream, 4 = North Atlantic Drift). This study focuses on both the spatial domain of WNAO defined here as the oceanic area bounded by 25–50°N and 60–85°W and North America’s East Coast. The yellow oval in the lower right panel represents the Sargasso Sea. The CAO shown in the top right panel is a NASA Worldview image (<https://worldview.earthdata.nasa.gov>) from 22 January 2019.

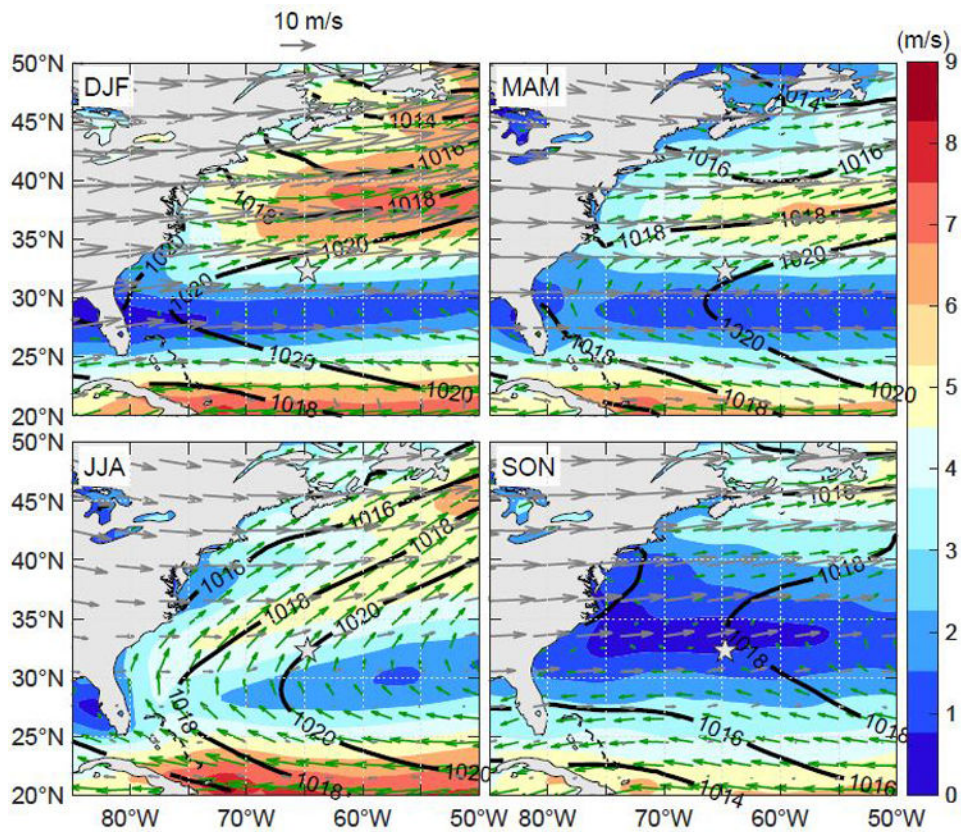


Figure 2. Seasonal meteorological fields taken from the Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) (Gelaro et al., 2017) for 2014 – 2018. Black contours represent sea level pressure (hPa), colored contours and green arrows are the magnitude and direction of near-surface winds at 975 hPa, respectively, and gray arrows are 700 hPa winds. December – January – February (DJF), March – April – May (MAM), June – July – August (JJA), and September – October – November (SON). The location of Bermuda is represented by the gray star.

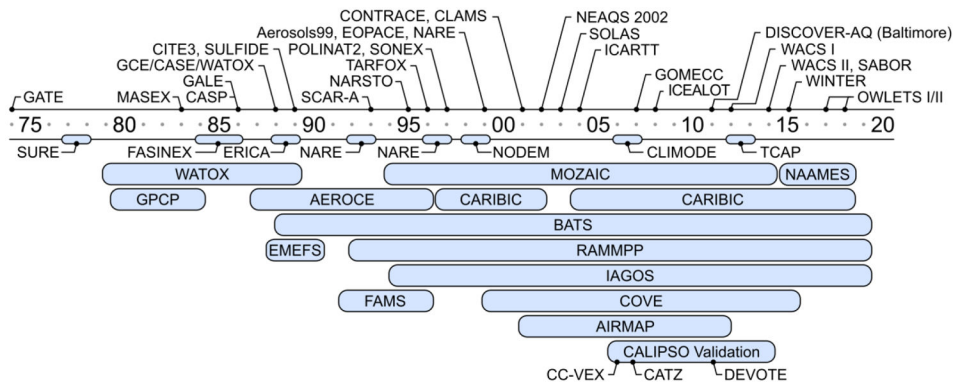


Figure 3. Timeline (1974 – 2019) of major field campaigns and long-term monitoring programs with measurements conducted in the extended WNAO region, which includes Bermuda and the East Coast of North America. NARSTO refers to the NARSTO-Northeast study. Note that some measurement efforts from Section 3 are not included here as they did not have a formal name.

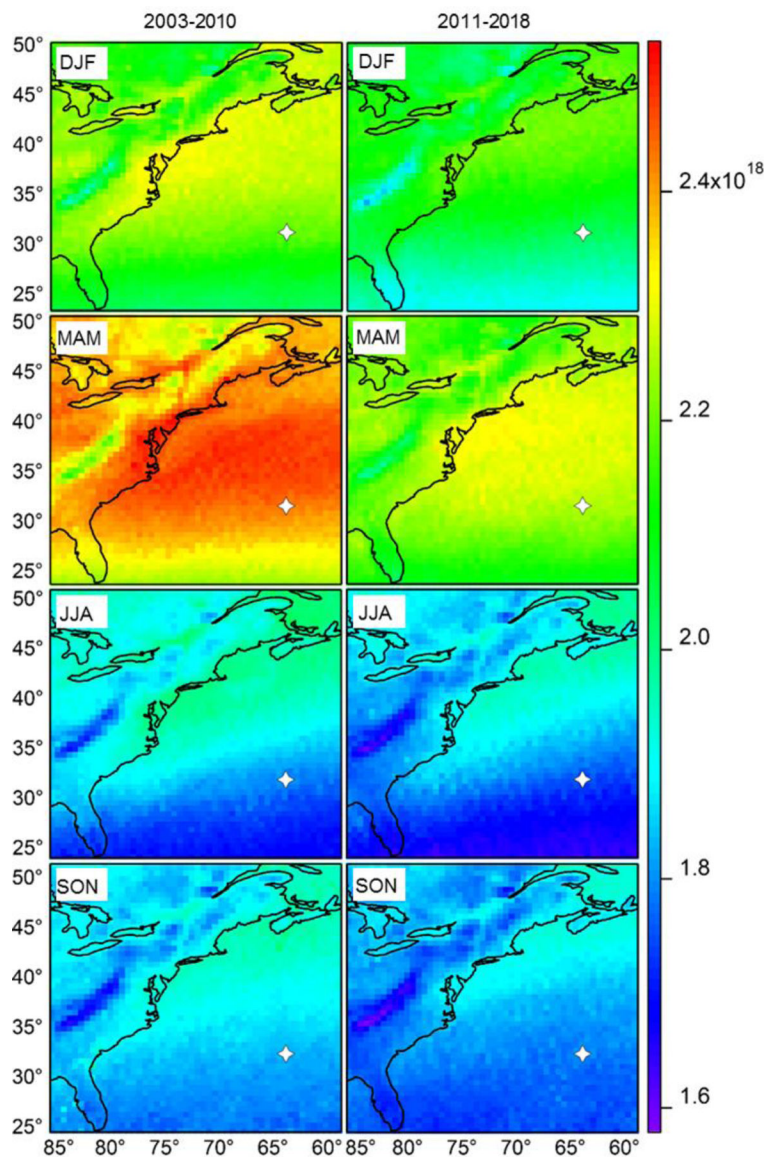


Figure 4. Seasonal maps of columnar carbon monoxide (CO) concentration (molecules cm^{-2}) from the Atmospheric Infrared Sounder (AIRS) over the extended WNAO region, including the eastern part of North America, categorized into two time periods (2003 – 2010 and 2011 – 2018). We specifically rely on the AIRS/Aqua L2 Standard Physical Retrieval (AIRS-only) V006 product at 50×50 km resolution (<https://search.earthdata.nasa.gov>). The gray marker represents Bermuda.

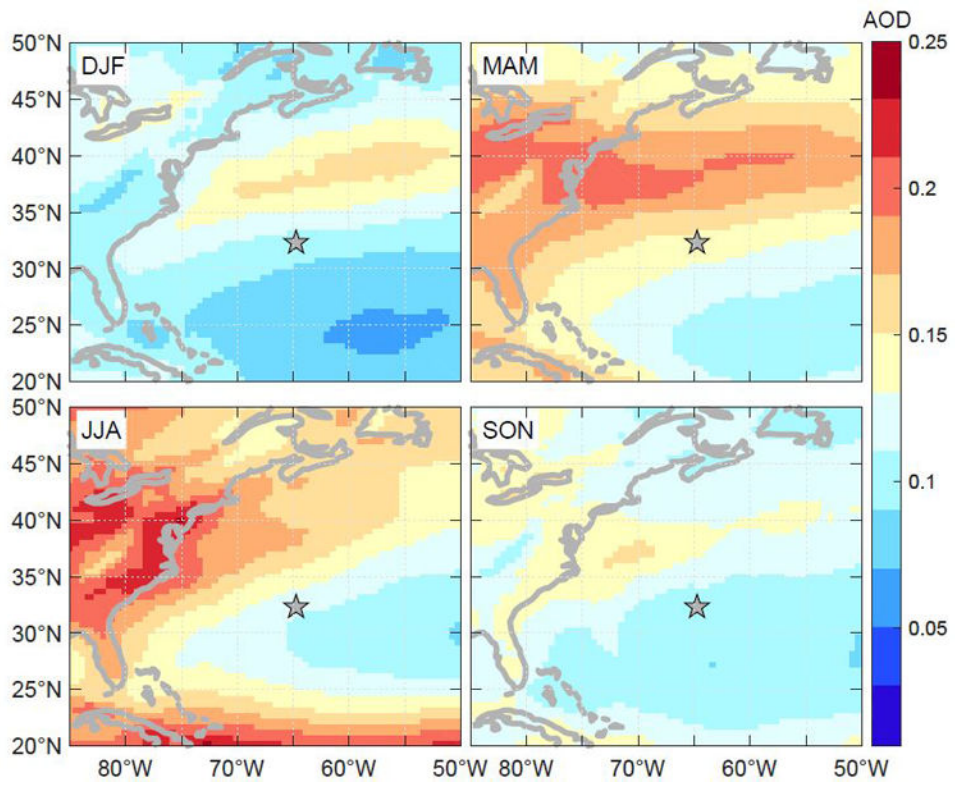


Figure 5.

As in Figure 2 but for aerosol optical depth (AOD) at 550 nm. Note that the MODIS AOD data were assimilated in MERRA-2 (Randles et al., 2017).

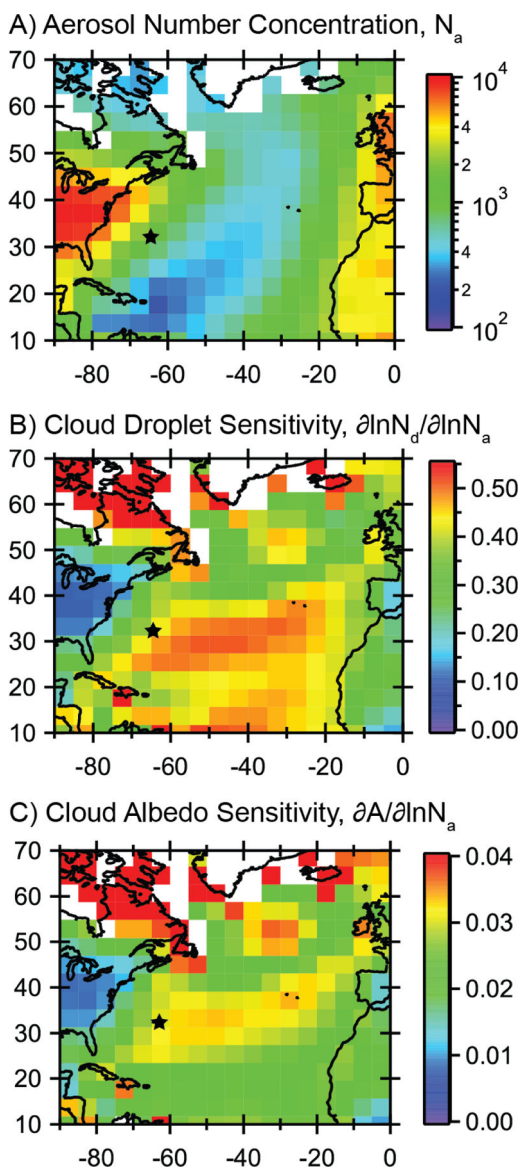


Figure 6. NASA Global Modelling Initiative (GMI) chemical transport model simulation results for (a) annual mean aerosol number (N_a), (b) logarithmic sensitivity of cloud droplet number concentration (N_d) to N_a , and (c) semi-logarithmic sensitivity of cloud albedo (A) to N_a . These results are from a single, climatological year (March 1997 to February 1998). The black marker represents Bermuda. Figure adapted from Moore et al. (2013), and used here under a CC Attribution 3.0 License.

Table 1.

Number of peer-reviewed publication relevant to atmospheric research for the WNAO region categorized based on topics and campaign/program. Gas/Aerosol/Wet Dep = studies examining any aspect of gas-phase, aerosol, or wet deposition properties, respectively, such as composition; Clouds = studies examining cloud/storm characteristics; ACI = aerosol-cloud interactions; Air-Sea = air-sea interactions; Dev't/Validation = studies examining any aspect of a new method, model, or instrument and/or validation studies between observations and models or retrievals; Met = studies with an extensive focus on meteorology and air movement. Rows showing multiple campaigns (e.g., "ICARTT/NEAQS 2002") indicate studies using data from each of the listed campaigns. "Bermuda" in some cases contains studies linked to other categories (i.e., AEROCE), while some AIRMAP studies fall into other campaigns (i.e., NEAQS 2002, ICARTT). IAGOS includes works from CARIBIC and MOZAIC. "Misc" includes studies not fitting under the other campaign/program names. Some publications addressed multiple topics and were therefore counted in multiple categories.

Campaign	Gas	Aerosol	Wet Dep	Clouds	ACI	Air-Sea	Dev't/Val	Met
Aerosols99	2	6	0	0	0	0	0	0
AIRMAP	12	7	0	0	0	0	2	3
Bermuda	38	59	37	10	4	32	22	6
CASP	0	0	0	1	0	0	0	0
CITE 3	6	1	0	0	0	0	1	1
CLAMS	1	10	0	0	0	0	11	0
CLIMODE	0	0	0	0	0	1	0	0
CONTRACE	1	0	0	0	0	0	0	0
COVE	0	0	0	0	0	0	9	0
DISCOVER-AQ	19	7	1	0	0	0	15	2
EMEFS	2	1	1	0	0	0	1	0
EOPACE	0	2	0	0	0	0	0	0
ERICA	0	0	0	4	0	3	0	6
FAMS	3	7	8	0	0	0	2	0
FASINEX	0	0	0	5	0	5	0	5
GALE	0	0	0	11	0	13	4	14
GCE/CASE/WATOX	14	15	6	0	0	2	2	1
GOMECC	3	0	0	0	0	3	0	0
GPCP	0	0	1	0	0	0	0	0
IAGOS/CARIBIC/MOZAIC	11	3	0	1	1	0	1	1
ICARTT	53	32	1	0	5	0	36	7
ICARTT/NEAQS 2002	2	0	0	0	0	0	1	1
ICEALOT	0	3	0	0	0	0	1	0
ICEALOT/WACS/NAAMES	0	1	0	0	0	0	0	0
MASEX	0	0	0	4	0	2	4	4
Misc	31	57	20	52	8	26	43	34
NAAMES	1	2	0	0	2	1	0	0
NARE	32	5	0	0	2	0	12	9

Campaign	Gas	Aerosol	Wet Dep	Clouds	ACI	Air-Sea	Dev't/Val	Met
NARSTO-Northeast	2	0	0	0	0	0	0	2
NEAQS 2002	10	5	0	0	0	0	2	0
NODEM	0	1	0	0	0	0	0	0
OWLETS	4	1	0	0	0	0	3	0
RAMMPP	6	5	0	0	0	0	2	1
SABOR	0	2	0	0	0	0	3	0
SCAR-A	0	2	0	0	0	0	1	0
SCAR-A/TARFOX	0	0	0	0	0	0	1	0
SOLAS	0	0	0	0	1	0	0	0
SONEX/POLINAT 2	8	7	0	0	0	0	0	2
SURE	3	3	0	0	0	0	0	0
TARFOX	4	16	0	0	0	0	11	0
TCAP	0	10	0	2	1	0	10	1
WACS	0	6	0	0	0	4	0	0
WACS II	0	1	0	0	0	1	0	0
WATOX	12	11	0	0	0	0	2	2
WINTER	6	6	0	0	0	0	3	0
Total	287	295	77	90	24	93	207	102
Relative Percent	24%	25%	7%	8%	2%	8%	18%	9%