Arctic Reactive Bromine Events Occur in Two Distinct Sets of Environmental Conditions: A Statistical Analysis of 6 Years of Observations

William F. Swanson1, Kelly A. Graham2, John W. Halfacre3, Christopher D. Holmes2, Paul B. Shepson4,5, and William R. Simpson1

1Department of Chemistry and Biochemistry and Geophysical Institute, University of Alaska Fairbanks, Fairbanks, AK, USA, 2Department of Earth, Ocean and Atmospheric Science, Florida State University, Tallahassee, FL, USA, 3Department of Chemistry, University of York, York, UK, 4Department of Chemistry, Purdue University, West Lafayette, IN, USA, 5School of Marine and Atmospheric Sciences, Stony Brook University, Stony Brook, NY, USA

Abstract Tropospheric bromine radicals in the Arctic efficiently remove ambient ozone and oxidize gaseous elemental mercury. Ground-based bromine monoxide (BrO) observations from the Arctic Ocean and Utqiâgteg (formerly Barrow) are combined with Modern Era Retrospective Analysis for Research and Applications version 2 reanalysis meteorological fields to determine how BrO varies with environmental conditions. The mean seasonal BrO abundance varies from year to year ($p < 0.001$), while regional variance in mean BrO is not statistically significant ($p > 0.11$). Principal component analysis derived three important principal components from the environmental data set. The third principal component explains the most variance in BrO and is correlated with low ozone and cold temperatures. This principal component is consistent with high BrO during ozone depletion events at cold temperatures and can work concurrently with each of the other two principal components to generate two distinct environmental types of high BrO events. The first principal component consists of a less-stable, thick, mixed layer and low atmospheric pressure and is consistent with observations of high BrO in low-pressure systems (e.g., storms). The second principal component consists of cold and stable conditions and is consistent with high BrO under surface-based temperature inversions. Our principal component regression model predicted the both the vertical column density of BrO in the lowest 2 km of the troposphere ($R = 0.45$) and the vertical column density of BrO in the lowest 200 m ($R = 0.54$). This statistical description of two types of reactive bromine events may help to harmonize space-based and ground-based observations.

1. Introduction

Enhanced levels of tropospheric reactive bromine radicals (Abbatt et al., 2012; Simpson et al., 2015) are observed by satellites each spring over the Arctic Ocean (Chance, 1998; Richter et al., 1998; Wagner & Platt, 1998). These high concentrations of reactive bromine in the Arctic boundary layer, along with chemically associated reactive chlorine (Custard et al., 2017) and reactive iodine (Raso et al., 2017), can lead to virtually complete removal of tropospheric ozone (Barrie et al., 1988; Simpson et al., 2007b), oxidation of hydrocarbons (Gilman et al., 2010; Jobson et al., 1994), and increased oxidation and deposition of gaseous mercury (Schroeder et al., 1998; Steffen et al., 2008; Wang et al., 2019).

Reactive bromine can be released into the Arctic troposphere by the production of molecular bromine ($\text{Br}_2$) through heterogeneous chemistry on frozen saline surfaces containing bromide (Custard et al., 2017; Fan & Jacob, 1992; Pratt et al., 2013; Vogt et al., 1996; Wren et al., 2013). Gaseous $\text{Br}_2$ easily photolyzes in the presence of sunlight and the resulting bromine (Br) radicals react with ozone to form bromine monoxide (BrO) radicals. Remote sensing methods detect optical absorption of BrO, which can be used as a proxy for total reactive bromine (Chance, 1998; Choi et al., 2012; Richter et al., 1998; Theys et al., 2011; Wagner & Platt, 1998). Self-reaction of BrO, reaction of BrO with ClO (Custard et al., 2016), or heterogeneous chemical cycles all regenerate Br and simultaneously deplete ozone (Abbatt et al., 2012; Simpson et al., 2007b; Simpson et al., 2015; Wang et al., 2019). In this manuscript, we will refer to the release of reactive bromine into the atmosphere from heterogeneous chemistry on frozen saline surfaces containing bromide as reactive bromine recycling. In the absence of heterogeneous recycling, BrO levels
photolytically decay within hours (Platt & Hönninger, 2003); thus, the presence of BrO is only observed when recycling is efficient.

Reactive bromine recycling chemistry in polar regions may be affected by surface salinity, temperature, pH, frozen saline surface area, and ozone mixing ratio, as previously demonstrated in laboratory experiments (Halfacre et al., 2019; Oldridge & Abbatt, 2011; Oum et al., 1998; Wren et al., 2013) and in the environment (Pratt et al., 2013). Modeling studies have shown that when O₃ mole fractions are as low as a few nanomoles per mole, as happens during severe ozone depletion events (ODEs), reaction of Br and O₃ to BrO is hindered and Br can be the major species in the BrOₓ family consisting of Br and BrO (Cao et al., 2014; Simpson et al., 2007b; Simpson et al., 2017; Wang et al., 2019). Reducing available BrO hinders conversion to HOBr that can undergo both the heterogeneous and multiphase reactions that recycle bromine (Abbatt et al., 2012; Simpson et al., 2015) and slows conversion to Br. Additionally, nitrogen oxides can form the reservoir species BrONO₂ (Custard et al., 2015) and enhance daytime Br₂ (Wang & Pratt, 2017). The distribution of reactive bromine species in the troposphere can also be affected by meteorology.

Many associations between meteorology and reactive bromine have been reported, but the full nature of these associations remains unclear. For example, rising temperatures, melting snowpack, and rainfall may reduce surface area available for atmospheric interaction, hindering reactive bromine recycling (Burd et al., 2017). The end of the reactive bromine recycling season is also associated with rainfall (Burd et al., 2017). Other meteorological processes have also been shown to increase bromide conversion to BrO, such as high wind speeds (Choi et al., 2012; Jones et al., 2009; Zhao et al., 2017), surface inversions (Frieß et al., 2004; Koo et al., 2012; Peterson et al., 2015), conditions with high aerosol extinction (Frieß et al., 2011), cold temperatures (Pöhler et al., 2010), and increased contact time between air masses and sea ice (Peterson et al., 2016; Simpson et al., 2007a). There are potentially several physical and chemical processes that could explain these associations.

The relationship between wind speeds, bromine activation, and ozone is especially complex. Most ozone depletion events have been reported to occur in conditions with low wind speeds under strong atmospheric stability (Halfacre et al., 2014; Simpson et al., 2007b), which allows reactive bromine to accumulate at the surface, where it depletes ozone (Lehrer et al., 2004; Peterson et al., 2015).

However, there are some reports of elevated reactive bromine and ozone depletion at high wind speeds, particularly from satellite instruments (Begoin et al., 2010; Blechschmidt et al., 2016; Jones et al., 2009; Zhao et al., 2016). These high-wind bromine events have been attributed to reactive bromine recycling on sea salt aerosol (SSA) particles sourced from blowing snow particles that are lofted into the atmosphere by the wind (Huang et al., 2018; Yang et al., 2008).

Jones et al. (2009) therefore proposed two regimes of increased reactive bromine levels and associated ozone depletion events: one regime at high wind speeds based on their satellite observations and another regime at low wind speeds based on literature observations of ozone depletion under calmer conditions. High winds and turbulence may also increase surface ozone by mixing ozone-rich air from aloft (Moore et al., 2014), and ground-based observations gathered over several years found no clear influence of surface windspeed on BrO abundance (Halfacre et al., 2014; Peterson et al., 2015).

The observations of two different bromine activation regimes may result from disparate factors affecting reactive bromine recycling during different circumstances. These differences may be resolved by examining reactive bromine observations across multiple years and regions of the Arctic Ocean. Springtime reactive bromine enhancements have been observed across many different regions of the Arctic Ocean, including on aircraft transects (Koo et al., 2012), near the Alaskan coast (Simpson et al., 2017), in marginal ice zones (Jacobi et al., 2006), and across large areas of the Arctic simultaneously by space-based instruments (Chance, 1998; Richter et al., 1998; Salawitch et al., 2010; Sihler et al., 2012; Wagner & Platt, 1998). Space- and ground-based observations observed BrO-enriched airmasses with distinct edges (Choi et al., 2012; Simpson et al., 2017). Space-based BrO retrievals have comprehensive coverage, but separating tropospheric BrO from total column BrO is be challenging due to temporal changes in stratospheric column BrO (Salawitch et al., 2010; Sihler et al., 2012; Theys et al., 2011) and potential effects of clouds. Addressing regional differences requires long-term observations from the Arctic Ocean itself, while addressing both interannual and regional differences requires a large data set covering multiple years and regions of the Arctic Ocean.
In this study, we utilize data from the O-Buoy project (Knepp et al., 2010) and coastal Utqiaġvik (formerly Barrow) to directly compare ground-based BrO observations gathered in multiple regions of the Arctic across multiple years to determine whether significant differences in the distribution of reactive bromine exist between years or regions. The correlative relationships between chemical and meteorological variables were additionally assessed by performing a principal component analysis, identifying principal components, which are correlated with two separate types of reactive bromine event. These correlations were subsequently tested by prediction of BrO via principal component regression. Section 2 explains the data sources and analysis methods. Section 3 reports results from the statistical test, which are then interpreted and compared to past literature in section 4. Section 5 concludes the paper.

2. Data Sources and Methods

2.1. O-Buoy Chemical and Temperature Observations

The O-Buoy project deployed 15 floating autonomous platforms (O-Buoys) from 2009 to 2015 as a part of the National Science Foundation-funded Arctic Observing Network project (Knepp et al., 2010). Data selected for this analysis are from 7 of the 15 O-Buoys that observed BrO (Carlson et al., 2010) during 2011 to 2016 from early spring to the seasonal end date (Burd et al., 2017). All O-Buoy data are available at arcticdata.io (Simpson et al., ). The locations of these buoys during the spring reactive bromine season are shown in Figure 1. The buoys were generally deployed in two regions: the Beaufort Gyre and near the North Pole.

Hourly BrO measurements were retrieved using multiple axis differential optical absorption spectroscopy (MAX-DOAS) (Carlson et al., 2010; Hönninger & Platt, 2002; Peterson et al., 2015). Our MAX-DOAS instruments used a scan pattern of four to seven elevation angles (generally 1°, 2°, 3°, 5°, 10°, 20°, and 90° elevation angles, but some early O-Buoys used a subset of these angles) (Simpson et al., 2017). Vertical profiles from this scan pattern were retrieved with an optimal estimation inversion procedure adapted from Frieß et al. (2011), used in Peterson et al. (2015), and under analysis settings described in Simpson et al. (2017) using a the zenith scan at 90° as the reference spectrum. The retrieval method constrains the BrO concentration and aerosol extinction to be positive at all altitudes; therefore, all retrieved quantities are positive although they may be very small. Examination of averaging kernels (e.g., see Peterson et al., 2015) shows that this vertical profile of BrO is best represented by two quantities: the vertical column density of BrO in the lowest 200 m, which is referred to as BrO$_{\text{surf}}$, and the vertical column density of BrO in the lowest 2,000 m of the troposphere, which is referred to as BrO$_{\text{L,Col}}$ (Peterson et al., 2015). Units for these quantities are in molecules/cm$^2$, representing the number of molecules in a vertical column with base area of 1 cm$^2$ and height of 200 m for BrO$_{\text{surf}}$ or 2,000 m for BrO$_{\text{L,Col}}$. Aerosol extinction in the lowest 200 m and lower tropospheric aerosol optical thickness were retrieved from MAX-DOAS measurements of O$_3$ collisional dimer by methods similar to those in Frieß et al. (2011), Peterson et al. (2015), and Simpson et al. (2017). MAX-DOAS observations of slant column density of NO$_2$ were additionally used for the pollution screen detailed below. Further information on the methods is detailed in the metadata files at the data repository, arcticdata.io (Simpson, 2018; Simpson et al., ).

The number of hourly BrO observations from each O-Buoy deployment and the periods of observation are shown in supporting information Table S1. The O-Buoy observations included BrO$_{\text{L,Col}}$, BrO$_{\text{surf}}$, aerosol extinction in the lowest 200 m, NO$_2$ slant column density, surface temperature, and surface ozone mixing ratio (2B Technologies Model 206, as described by Halfacre et al. [2014]).

2.2. Utqiaġvik Chemical and Temperature Observations

A MAX-DOAS was deployed in 2012 at the Barrow Arctic Research Center (BARC), which is located at 156.6679°W, 71.3249°N near Utqiaġvik, AK (Simpson, 2018), with data available at the arcticdata.io repository (Simpson, 2018). The same retrieval method used for the O-Buoy instruments was used to retrieve BrO$_{\text{surf}}$, BrO$_{\text{L,Col}}$, aerosol extinction, and NO$_2$ slant column density. Utqiaġvik was defined as the third observation region for statistical analysis. The viewpoint on the MAX-DOAS at BARC was heated to prevent frosting, allowing it to start observations as soon as there was sufficient sunlight. During 2012, the MAX-DOAS data at BARC was compared with two O-Buoy-like Icander platforms (deployed on top of sea ice instead of within), showing measurements from various MAX-DOAS systems are comparable (Simpson et al., 2017). BARC MAX-DOAS data from 2012 to 2016 is added to the O-Buoy data for this
The number of hourly BrO observations during each year at BARC and the periods of observation are shown in the supporting information Table S1.

Ozone and temperature were measured in Utqiaġvik at the NOAA Barrow Atmospheric Baseline Observatory located 2.7 km from BARC at 156.6114°W, 71.3230°N. Hourly in situ ozone measurements (McClure-Begley et al., 2014) and temperature at 2 m (NOAA, 2018) were provided by the NOAA Earth System Research Laboratory.

2.3. Meteorological Reanalysis Data

The Modern Era Retrospective Analysis for Research and Applications version 2 (MERRA-2) is a meteorological reanalysis product provided by the National Aeronautics and Space Administration Global Modeling and Assimilation Office (Gelaro et al., 2017). We extract meteorological data along O-Buoy location tracks and at Utqiaġvik using spatial linear interpolation between MERRA-2 grid cells at 2° latitude by 2.5° longitude grid resolution. Interpolated sea level pressure, wind speed at 10 m, and potential temperature at heights of 2, 100, and 1,000 m were obtained in this way. The mixed layer height (as defined by the preferred estimate [Molod, 2014] using the bulk Richardson number with a critical value of 0.25 [Seibert et al., 2000]) was calculated from MERRA-2 using vertical profiles of virtual potential temperature and windspeed.

2.4. Variable Selection, Normalization, and Screening

Our goal is to understand the meteorological and environmental factors that influence hourly BrO_L,Tcol and BrO_surf measurements from MAX-DOAS instruments. BrO_L,Tcol and BrO_surf are treated as the dependent variables in this analysis. To probe the correlation of BrO with environmental variables in the Arctic spring, we used O-Buoy observations of aerosol extinction, surface ozone mixing ratio, and surface temperature at heights of 2, 100, and 1,000 m were obtained in this way. The mixed layer height (as defined by the preferred estimate [Molod, 2014] using the bulk Richardson number with a critical value of 0.25 [Seibert et al., 2000]) was calculated from MERRA-2 using vertical profiles of virtual potential temperature and windspeed.

These variables have been previously associated with BrO abundance. High aerosol extinction in the lowest 200 m (EXT) has been previously linked to elevated BrO near the surface at Utqiaġvik (Frieß et al., 2011).
Ozone mixing ratio was also considered for multiple reasons. The reaction of ozone with bromine radicals is the main source of BrO (Abbatt et al., 2012), and exposure of snowpack to increased levels of ozone can lead to the production of molecular bromine (Pratt et al., 2013). Ozone can also be an indicator of airmass history, such as high surface ozone due to the vertical mixing of ozone rich air from aloft to the surface (Moore et al., 2014). Surface temperature was included because subfreezing temperatures appear to be required for reactive bromine recycling (Burd et al., 2017; Peterson et al., 2015; Pöhler et al., 2010) and some past work indicated very cold temperatures may enhance bromine activation (Pöhler et al., 2010). Though O-Buoys measured SLP, MERRA-2 SLP was included here because it agrees closely with the O-Buoys (see supporting information Figure S1) and the higher precision allows for the hourly rate of change in pressure (ΔP1hr) to be calculated from MERRA-2 sea level pressure using equation (S1). MERRA-2 windspeed at 10 m above the surface (Vwind) was analyzed based on a proposed link between high BrO and high windspeed (Jones et al., 2009). Atmospheric stability affects the vertical distribution of BrO (Peterson et al., 2015). A larger (positive) difference in potential temperature between two altitudes corresponds to increased atmospheric stability between those altitudes. The potential temperature differential in the lowest 1,000 m was calculated using equation (S2) as the difference in MERRA-2 potential temperature from 1,000 to 2 m (ΔT1000m). A height of 1,000 m was chosen because ozone depletion events are usually confined to the lowest 1,000 m (Bottenheim et al., 2002; Salawitch et al., 2010). We chose to include temperature differential at another height to probe stability and mixing near the surface. A height of 100 m was chosen because strong inversions often form at low heights over the Arctic Ocean (Anderson & Neff, 2008). The potential temperature differential in the lowest 100 m was calculated using equation (S3) as the difference in MERRA-2 potential temperature from 100 to 2 m (ΔT200m). Vertical mixing can occur more freely within the mixed layer. Several methods of calculating the mixed layer height were examined, and the bulk Richardson estimate of mixed layer height from MERRA-2 temperature and windspeed was selected for this analysis as detailed in section 2.3.

The hourly BrO observations and aerosol extinction in the lowest 200 m have non-Gaussian distributions, as shown in the left column of supporting information Figure S2. To normalize these variables to a near Gaussian distribution, a square root transformation was applied to BrOLTcol and BrOsurf and a natural logarithm transformation was applied to aerosol extinction. The near-Gaussian distributions of the normalized variables are shown in the right column of supporting information Figure S2. All variables including the transformed variables were then standardized to a mean of 0 and a standard deviation of 1 for principal components analysis. This standardization is done so that variables with different units such as pressure and extinction coefficient may be included in the same analysis despite having unequal variance.

To identify the correlation between elevated reactive bromine levels and local environmental variables, general conditions must be conducive to reactive bromine events and our observations of BrO must closely represent the total amount of reactive bromine in the troposphere. To meet these criteria, all data were screened for rainfall, observation date, ozone concentration, and pollution as follows.

**2.4.1. Rainfall Screen**

Burd et al. (2017) studied the termination of BrO recycling in the Arctic and found that BrO was absent during and after rainfall. Therefore, MERRA-2 was used to estimate liquid precipitation, calculated as the difference between MERRA-2 total precipitation and snowfall. This rainfall variable was used to remove time periods when BrO would not be present as recycling would have been hindered.

**2.4.2. Seasonal End Date Screen**

Reactive bromine chemistry is no longer observed after the seasonal end date. The seasonal end date is defined as the time at which BrO is below a certain threshold (1° dSCD is below 5 × 1013 molecules/cm²) and the 3-hr average of BrO does not rise above that threshold for the next 5 days. Only observations made from the beginning of observation until 5 days after the BrO seasonal end date were used in this analysis. The periods of observation for each year are shown in supporting information Table S1.

**2.4.3. Ozone Screen**

During severe boundary layer ozone depletion events when ozone falls below 1–2 nmol/mol, there is insufficient ozone to convert Br radicals to BrO, and BrO becomes a minor component of reactive bromine (Helmig et al., 2012; Simpson et al., 2007b, 2017; Wang et al., 2019). Under these extremely low ozone conditions, BrO no longer accurately represents the total amount of reactive bromine present. The 2B Technologies Model 206 ozone instruments used on the O-Buoys have an uncertainty of roughly
2 nmol/mol (Halfacre et al., 2014). Therefore, only observations when the ozone is measured above 2 nmol/mol are used in this analysis.

### 2.4.4. Pollution Screen

Only observations of reactive bromine in unpolluted airmasses are analyzed in this study. Nitrogen dioxide (NO₂) has a lifetime in the boundary layer of about 1 day (Wenig et al., 2003), and MAX-DOAS differential slant column densities of 2° above horizontal (2° dSCD) can therefore be used as a proxy for fresh anthropogenic pollution. All observations made when the measured NO₂ 2° dSCD was more than 5 x 10¹⁵ molecules/cm², or roughly three times the measurement error in the UV band, were removed from the data set.

### 2.5. Statistical Methods

The R Project computing environment version 3.3.1 was used in this analysis (Team R Core, 2018). The core environment contains the analysis of variance and principal component analysis functions used in this analysis. The PLS package was used to perform principal components regression (Mevik et al., 2019).

#### 2.5.1 Analysis of Variance

Analysis of variance (ANOVA) was used to determine whether BrO observations from different years or regions were statistically significantly different from each other (Chambers & Hastie, 1992). We performed a two-tailed ANOVA on both BrOLTcol and BrOsurf with two statistical factors: year of sampling and regions of sampling. The three regions (defined in Figure 1) are the Beaufort Gyre, North Pole, and Utqiagvik. This test assumes that the variables have Gaussian distribution, equal variance, and independently sampled observations in each case. The BrO data was normalized by the methods detailed in section 2.4 to transform the data to have a nearly Gaussian distribution, shown in the supporting information Figure S2. To minimize autocorrelation and satisfy the independence assumption for the ANOVA, the hourly BrO observations were averaged to 24-hr daily BrO averages.

#### 2.5.2. Principal Components Analysis

Principal components analysis (PCA) is a statistical procedure that can be used to identify patterns among variables that explain the most variance within a data set (Mardia et al., 1979). We used it to determine how the meteorological and chemical variables listed above associate with each other. For this analysis, each variable must have a Gaussian distribution or be transformed (see section 2.4) to be near Gaussian. Each variable is then standardized by subtracting the mean and dividing by the standard deviation. Standardization allows the PCA to compare the contribution of each variable on an equal footing, as they all have equal variance after transformation. If the variables were not standardized, the high variance of one variable might outweigh the contributions to variance from other variables. Hourly observations of variables are used in the PCA, as this statistical procedure does not require the independence of cases that an ANOVA does. Standardizing the distributions aids comparison between variables with different units and ensures that each variable contributes equally to the total variance of the data set. Decomposition of the resulting correlation matrix results in orthogonal principal components (PCs) equal to the number of original variables. Each PC with a variance greater than unity explains more normalized variance within the data set than any single variable. To determine how robust the PCA results are, a sensitivity test was performed by running a second PCA with only meteorological variables, excluding ozone and extinction. The loadings of each PC describe the correlation of each constituent variable to the entire PC. The PCs are often called empirical orthogonal factors, and the term “factor” is used henceforth to refer to the meaning of a PC in the context of this specific data set. Principal component scores for each leading PC with variance greater than unity were calculated by multiplying the timeseries of each variable value by the variable loading and repeating for each variable in the PCA.

#### 2.5.3. Principal Component Linear Regression

Principal components regression (PCR) fits a variable of interest to the leading principal component scores (Wilks, 2011). Ordinary least squares linear regression is used to relate the timeseries of the variable of interest to the timeseries of PCs with a variance greater than unity or the first three PCs in this data set (Mevik et al., 2019). This process was performed once for BrOLTcol and again for BrOsurf to yield two equations to predict standardized BrOLTcol and BrOsurf based on linear combinations of each variable used in the PCA multiplied by an empirically derived coefficient.
3. Results

3.1. Data Set Characterization

The application of data screens narrows 6 years of spring O-Buoy observations to 3,887 hourly measurements of BrO_LTcol, BrO_surf, and simultaneous meteorology (Table 1). The majority of screened O-Buoy data comes from O-Buoy 2 in 2011, O-Buoy 4 in 2012, O-Buoy 10 in 2014, and O-Buoy 11 in 2015. A timeseries of O-Buoy 10 BrO observations is shown in Figure 5 in blue. Full timeseries of BrO observations on O-Buoy 2, 4, and 11 and for all 5 years at Utqiaġvik are shown in the supporting information Figures S3–S10. The largest rejection of data was caused by a lack of concurrent ozone observations on O-Buoy 2, with roughly half of BrO_LTcol and BrO_surf observations omitted because of O-Buoy ozone instrument failure on other buoys after deployment. The rainfall screen cut roughly a quarter of the data points. The pollution screen eliminates roughly 5% of the data at Utqiaġvik and a single hourly observation from the O-Buoys. Omitting times at which some variable other than BrO or ozone was missing cuts another 5% of observations. A third of screened BrO_surf observations were eliminated because there must be both valid BrO_surf and BrO_LTcol.

The means and ranges of 1 standard deviation for each variable are shown in Table 2. The average surface ozone mixing ratio of 21 nmol/mol is at the low end of the global background surface ozone (Vingarzan, 2004). Tropospheric ODEs are common during the Arctic spring (Barrie et al., 1988; Simpson et al., 2007b) and cause a large standard deviation of ozone. The average surface temperature in this data set is well below freezing due to the inclusion of a rainfall screen and ending observations based upon the seasonal end date. The average sea level pressure of 1,020 hPa is normal for the Arctic from February to April (Przybylak & Wyszy, 2013). Surface windspeed of 5.6 ± 2.8 m/s is slightly higher than historical windspeed distributions of 2.5–4 m/s observed in the North Slope and Beaufort Sea region in May and June (Segall & Zhang, 2012), possibly because we included April observations when winds speeds are greater. The average Richardson mixed layer height is 330 m, with a standard deviation of 170 m. This average mixed layer height is on the high end of the range of boundary layer heights observed by radiosondes launched twice daily at Utqiaġvik during the OASIS campaign in 2009 (Boylan et al., 2014). The higher mixed layer height may be due to thicker boundary layers later in the spring season, as the OASIS campaign ended on April 15 while we continue observations in June. Average pressure change across a large set of observations should be equal to zero. The average pressure change, 0.3 hPa/hr, is within 1 standard deviation of zero. Potential temperature increased with altitude at a mean rate of 11.5 K in the lowest kilometer compared to the standard dry adiabatic potential temperature lapse rate of 0 K/km. Boundary layers over snow and ice tend toward being more stable, and temperature inversions commonly form (Anderson & Neff, 2008). Strong temperature inversions may be associated with an increase in potential temperature from the surface to 1,000 m. Our potential temperature differential in the lowest 1,000 m is 11.5 K, indicating a stable lowest 1,000 m. The average increase in potential temperature indicates a boundary layer that is stable on average. The average potential temperature differential between 0 and 100 m was 0.3 K, indicating that the lowest part of the atmosphere is also stable. The distribution of potential temperature differential in the lowest 100 m shown in the supporting information Figure S11 is skewed to the right, with most potential temperature differentials clustered within 1 K of zero and a small fraction (574 of 3,887) of potential temperature differentials ranging from 1 to 6 K. Less than half (1,913 of 3,887) of the temperature differentials are negative, meaning unstable, but none of the differentials drop below −1 K. Most (1,405 of 1,913) of the negative temperature differential cases occurred at Utqiaġvik and may be due to MERRA-2 aggregation error in which the grid cell containing Utqiaġvik also includes open leads that may be present along the coastline west of Utqiaġvik (Braham et al., 1980). Open sea ice leads can lead to convective mixing (Moore et al., 2014). The presence of open ocean with a temperature near freezing can expose relatively warm water to subzero air in the Arctic spring; if the sea surface is warmer than the air above it, convective mixing can occur, possibly contributing to the negative potential temperature differentials seen in this data set.

3.2. Regional and Interannual Variability

A two-tailed analysis of variance tested whether there are significant regional or interannual differences in mean BrO in the screened daily averaged BrO data set based on two statistical factors of year and region. Hourly observations were screened for rainfall, ozone, and pollution before daily averaging. The screened daily averaged data set contained 611 days of BrO_surf observations and 548 days of BrO_LTcol observations. Figure 2 reports the 25th, 50th (median), and 75th percentiles of daily BrO values by year and region.
Exact values and sample sizes are shown in Tables S3 and S4. Across years, median daily BrO differed by nearly a factor of 4 for BrOLTcol ((5.6 to 21.5) × 10^{12} molecules/cm^2) and more than a factor of 2 for BrOsurf ((3.2 to 8.0) × 10^{12} molecules/cm^2). The ANOVA revealed that interannual variations in mean BrO are significant (p < 0.001) for both BrOLTcol and BrOsurf. Regionally, the median BrO in the Beaufort Gyre region is roughly half the median BrO in the North Pole and Utqiagvik regions, but these differences have less statistical significance (p = 0.16 for BrOLTcol and p = 0.11 for BrOsurf). The total days of observation are similar for each year but unequal for each region, with roughly 50 days of coverage in the North Pole, roughly 150 days in the Beaufort Gyre, and almost 400 days at Utqiagvik.

3.3. PCA Loadings

We combined the nine chemical and meteorological variables discussed in section 2.4, excluding all measurements of BrO, to create a PCA data set. Three PCs have variances greater than 1, meaning that they explain more variability than any individual variable alone. Principal component one (PC1) has a normalized variance of 2.17; principal component two (PC2) has a normalized variance of 1.78; and principal component three (PC3) has a normalized variance of 1.43. These PCs meet the North et al. (1982) criteria for independence because the minimum difference in variance between PCs (0.347) is much greater than the sampling error (0.033). The loadings of these three PCs are plotted in Figure 3 and appear in the supporting information Table S2. Together, they explain 60% of the variance in the data set. Principal components 4–9 explain variance less than unity.

PC1 consists mainly of thick mixed layer height and decreased stability in the lowest 100 m, with some connection to increased surface wind speeds, decreased sea level pressure, decreased stability in the lowest 1,000 m, and increased aerosol extinction. PC1 has minimal connection with pressure change, ozone, or temperature. PC2 consists mainly of increased stability in the lowest 1,000 m, high surface wind, and cold
temperatures, with some connection with increased ozone, sea level pressure, aerosol extinction, and falling pressure. PC2 has minimal connection with mixed layer height or stability in the lowest 100 m. PC3 consists mainly of cold temperatures and low ozone, with some connection with increased sea level pressure, aerosol extinction, and increasing pressure and minimal connection with mixed layer height or stability in the lowest 1,000 m. Figure S12 shows the results of a PCA where the PCA excludes ozone and extinction. All three PCs in the main PCA resemble their counterparts in the sensitivity test. Section 4 will interpret and discuss the principal components.

3.4. Prediction of BrO

We use the first three PCs to predict BrO_{LTcol} and BrO_{surf} via the PCR method. These components each explain a normalized variance of greater than unity in the PCA, and the PCR predictive model shows that each component explains an amount of variance in BrO_{LTcol} or BrO_{surf}. PC3 is most influential for predicting both BrO_{LTcol} and BrO_{surf}, explaining 18.0% and 22.9% of their respective variances. PC1 explains 10.2% of BrO_{LTcol} variance but still explains 4.8% BrO_{surf} variance. Conversely, PC2 accounts for only 0.6% of the variance and 9.8% of BrO_{surf} variance. The fourth–ninth PCR components combined explain only 6% of variance in BrO_{LTcol} and 2% of variance in BrO_{surf}. Both BrO measurements are most strongly associated with PC3. PC1 is more strongly associated with BrO_{LTcol} while PC2 is more strongly associated with BrO_{surf}. The results of the PCR are captured in equations (S4), which are used to predict BrO_{LTcol} and BrO_{surf}.

Figure 4 shows predictions of BrO_{LTcol} and BrO_{surf} from the fitted PCR model versus observations. For BrO_{LTcol}, the correlation coefficient between the predictions and observations is \( R = 0.45 \) and 1,722 of 3,778 values are within average observational error. The correlation coefficient between the BrO_{surf} predictions and observations is stronger \( (R = 0.54) \) and 1,862 of 3,778 values are within average observational error. The mean bias in the predictions is \( 2.0 \times 10^{12} \) molecules/cm\(^2\) below observations for BrO_{LTcol} and 0.6 \times 10^{-12} \) molecules/cm\(^2\) below observations for BrO_{surf}. Most of the extremely high BrO observations occur when predicted BrO is above the median, meaning that the statistical model has some ability to predict high BrO but not to the extent observed. The model has slightly higher nonparametric Spearman rank correlation \( (\rho) \) coefficients \( (\rho = 0.56\) and 0.61 \) for BrO_{LTcol} and BrO_{surf}, respectively \) than linear correlation coefficients \( (R = 0.45\) and 0.56 \) for BrO_{LTcol} and BrO_{surf}, respectively \). The nonparametric Spearman rank correlation coefficient is insensitive to the magnitudes of the highest values, further confirming the model’s ability to qualitatively predict the periods of high BrO. Figure 5 shows a sample timeseries of PCR predictions and observations of BrO_{LTcol} from O-Buoy 10. This timeseries covers observations from Spring 2014 in the
beaufort sea region. this timeseries demonstrates that episodes of high and low brO are well predicted but that the range of brO extremes is not well predicted.

4. discussion

4.1. interannual variation in arctic brO

The ANOVA found significant interannual variation in daily averaged Arctic springtime BrO at a 99% confidence level ($p < 0.001$). The significant interannual variation within this data set can be seen in the varying distributions of both BrO data products in Figure 2. This agrees with a previous study of satellite-derived data that found that the strength of BrO events varies from year to year (Hollwedel et al., 2004). This finding is also consistent with a study of ozone depletion events at Utqiaġvik showing significant interannual variability in the frequency of low ozone (<10 ppb) at events (Oltmans et al., 2012).

Several processes contribute to interannual variation in mean BrO. Changes in transport may expose observed airmasses to frozen saline surfaces that are more effective at recycling reactive bromine, such as surface snow over first-year ice with an ionic composition differing from surface snow over multiyear ice (Krnavek et al., 2012). Peterson et al. (2016) examined Utqiaġvik observations from 2008, 2009, 2012, and 2013 and found that annual averages of coastal BrO$_{L, Tcol}$ were highly correlated with annually averaged first-year ice contact, which was attributed to interannual variability in transport. The chemical composition of surface snow atop ice or tundra varies widely on small spatial scales (Jacobi et al., 2012; Krnavek et al., 2012), and low salinity snow surfaces can lead to effective halogen activation (Custard et al., 2017; Pratt et al., 2013; Raso et al., 2017). Therefore, lengthy changes in the synoptic scale weather patterns that transport air masses over snow and sea ice of widely varying composition could lead to significant variation on a daily or monthly timescale (Koo et al., 2014).
Supporting information Figure S13 shows that the highest mean BrOLTcol and BrOsurf occurs in the years with smallest minimum sea ice extent in the previous year, but the correlation between BrO and sea ice is not significant ($R = -0.59$ for BrOLTcol [$p = 0.21$] and $R = -0.49$ for BrOsurf [$p = 0.33$]). Further field studies in future years may be able to make more substantial conclusions about the relationship between annual BrO and sea ice.

Significant interannual variation in BrO could additionally be related to variation in surface temperature. Arctic surface air temperature shows high decadal and interannual variability (Van Der Linden et al., 2017). Surface temperature increases lead to early snowmelt in spring, and melted snow hinders reactive bromine recycling (Burd et al., 2017). The loadings of PC2 and PC3 in Figure 3 show that cold temperatures are correlated with increased BrOLTcol and BrOsurf. Interannual temperature variation could contribute to the interannual variation in reactive bromine found here. However, supporting information Figure S14 shows that there is no clear linear relationship between surface temperature and hourly BrOLTcol or BrOsurf in our data set. The relationship between temperature and BrO is explored further in section 4.3.

Figure 4. Two-dimensional histograms showing density of predicted BrO versus observed BrO, with (a) BrOLTcol shown on top sorted into square bins of 0.2 and (b) BrOsurf on the bottom sorted into square bins of 0.05. All units are in molecules/cm$^2$. The 1:1 line is drawn in the center in black, with a margin of the average observational error plotted in dashed black lines around the central 1:1 line.
4.2. Spatial Variation of BrO Across the Arctic Ocean

Previous analyses of Arctic data sets have examined spatial variability in ozone depletion caused by reactive bromine chemistry (Halfacre et al., 2014) as well as the correlation between BrO and airmass time in first year ice areas (Peterson et al., 2016). In this data set, the Beaufort Gyre experienced lower median BrOLTcol and BrOsurf by a factor of 2 than either the North Pole region or Utqiaġvik (see Figure 2). These probabilities do not rise to the 95% confidence level within this data set ($p = 0.11$ for BrOsurf, $p = 0.16$ for BrOLTcol). Additionally, the unequal distributions of data coverage in each region reduces our confidence in ANOVA conclusions on regional variation. More evidence is needed to determine if the modest regional differences found in these measurements are meaningful.

One source of regional variation could be terrestrial influences, such as transported plumes of pollution or smoke. Our data set was gathered over the Arctic Ocean and at Utqiaġvik, which is subject to prevailing winds coming from the Arctic Ocean (Peterson et al., 2016). Our pollution screen also acts to filter out polluted air masses arriving from inland at Utqiaġvik. Airmasses remain in the Arctic above 70°N for 1–2 weeks (Stohl, 2006) where local pollution is minimal. We conclude from our results that additional observations are needed to determine if differences in mean BrO abundances across different regions of the Arctic Ocean are present.

4.3. The Third Principal Component Describes an Ozone Depletion Chemical Factor

PC3 explains the most variance in BrO in the PCR despite being the third most important driver of variance in the PCA data set. The third PC consists of low ozone mixing ratios and cold surface temperatures, resembling a typical Arctic springtime ozone depletion event. Ozone depletion events have been consistently observed in conjunction with elevated bromine monoxide due to the reaction of ozone with reactive halogens (Fan & Jacob, 1992; Molina & Rowland, 1974; Simpson et al., 2007b), and the rate of ozone depletion has been shown to be kinetically consistent with observed Br atom concentrations (Wang et al., 2019). The low ozone loadings of PC3 and the fact that PC3 explains most variance within BrOLTcol and BrOsurf...
support the idea that this factor largely describes the chemical destruction of ozone with increasing reactive bromine. At very low ozone levels, BrO cannot be formed (Simpson et al., 2015). However, by screening out observations with ozone below 2 nmol/mol, this aspect of the chemical relationship between BrO and ozone is excluded from the data set.

Surface temperature is an important indicator of meteorological conditions, but it can also impact chemical recycling. Extremely cold temperatures (<252 K) can precipitate salts such as hydrohalite (Linke, 1965), reducing the availability of chloride (Sjostedt & Abbatt, 2008) and allowing for increased formation of Br2 within the snowpack (Custard et al., 2017). Pöhler et al. (2010) observed increased BrO at temperatures below −15 °C. Reactive bromine recycling has been observed at temperatures up to −5 °C over a longer (five spring seasons) data set (Burd et al., 2017), but temperatures above freezing can hinder reactive bromine recycling. A similar relationship is seen in the supporting information Figure S14, where the BrO observations approach zero above freezing.

Variables besides ozone and temperature have relatively weak loadings (<0.4) within PC3. The connections to high pressure, increasing hourly rate of pressure change and low winds resemble quiescent synoptic conditions without low-pressure systems or cyclones. When ozone and extinction are eliminated from the PCA as in the sensitivity test (Figure S12), the chemical aspect of PC3 disappears but the high-pressure meteorological conditions remain, with the rate of pressure change increasing. The loadings for wind speed and potential temperature differential in the lowest 100 m also increase in magnitude. These variables indicate higher pressures and a slightly calmer conditions when this chemical factor is working concurrently with the other two PCs. Increased vertical mixing of ozone rich air in a low-pressure system would decrease the relative importance of chemical depletion of ozone. PC3 is a chemical factor that largely tracks the chemical relationship between BrO, ozone, and temperature during calmer meteorological conditions.

4.4. The First Principal Component Describes a Low Pressure, Less Stable Meteorological Factor

PC1 consists of low sea level pressure, decreased stability in the lowest 100 and 1,000 m, and increased mixing height. The highest loadings in PC1 are all characteristic of low-pressure meteorological systems that can increase vertical mixing in the boundary layer. Arctic low-pressure systems have been observed in conjunction with enhanced BrO (Choi et al., 2012; Jones et al., 2010; Sihler et al., 2012). Peterson et al. (2015) observed the highest total BrO_Ltcol when BrO was distributed throughout the troposphere instead of concentrated in the lowest 200 m of the atmosphere. PC1 has connection to a less stable lowest 100 and 1,000 m based on the temperature differentials. This lowered stability and increased vertical mixing could act to vertically distribute aerosol, allowing for recycling of bromide to reactive bromine on aerosol surfaces throughout the lower troposphere. When ozone and extinction are removed from PCA analysis as in Figure S12, PC1 remains the same as in Figure 3. The first PC primarily describes a meteorological factor with low pressure systems and a thick mixed layer where aerosols containing the reactive bromine precursor bromide can be vertically distributed throughout the lower troposphere.

4.5. The Second Principal Component Describes a Meteorological Factor With Cold Stable Inversions

PC2 consists of cold surface temperatures, high winds, and increased stability in the lowest 1,000 m. Increased stability in the lowest 1,000 m may indicate presence of a temperature inversion. The connection to variables besides temperature and stability expands the understanding of this meteorological factor beyond a simple temperature inversion. The correlation with wind speed could be an indicator of the importance of wind pumping, which plays an important role in transporting reactive bromine from the snowpack to the surface layer (Thomas et al., 2011). The weaker correlation with low hourly rate of pressure change indicates that the pressure is dropping slightly, which combined with the high winds may indicate oncoming low-pressure systems. Increased ozone is effective at increased BrO recycling. PC2 in Figure 3 shows minimal variation from PC2 in Figure S12 where ozone and extinction are removed from PCA analysis. Taken as a whole, the second PC primarily describes a meteorological factor with cold surface temperatures, high winds, and a stable mixed layer that allows for efficient recycling of BrO on snowpack surfaces containing bromide.
4.6. The Three Principal Components Work Concurrently to Describe Two Types of Bromine Events

The chemical factor (PC3) consists of low ozone and cold temperatures and explains the largest amount of variation in both BrO_LTCol and BrO_surf. Both chemical and meteorological factors can work at the same time to enable more efficient recycling of BrO. While the PCs derived from the PCA are orthogonal to each other, they can contribute concurrently to predict two distinct types of high BrO event.

Variance in BrO_LTCol is mainly explained by the chemical factor (PC3) and the low-pressure vertically distributed meteorological factor (PC1). This meteorological factor (PC1) explains more than double the variance of BrO_LTCol than BrO_surf. The most important aspects of the meteorological factor are vertical mixing under a thick mixed layer in a low-pressure system. Choi et al. (2018) found that satellite detected BrO increases at higher wind speeds driven by pressure gradients. Reactive bromine events caused by concurrent contributions from PC1 and PC3 would agree with space-based observations of high BrO in low-pressure systems (Blechschmidt et al., 2016; Jones et al., 2009; Zhao et al., 2017).

We can approximate environmental conditions during an event when PC1 and PC3 work concurrently at comparable strength by adding the loadings from each PC. The resulting high BrO_LTCol event occurs under environmental conditions with higher extinction, a thick boundary layer, low ozone, and lower stability in the lowest 100 m. Aerosol extinction is correlated with enhanced BrO from ground-based MAX-DOAS (Frieß et al., 2011) and satellites (Choi et al., 2012). Reactive bromine recycling in this event could be enhanced by heterogeneous chemistry on SSA sourced from blowing snow (Kirpes et al., 2018; Yang et al., 2008). The influence of the low-pressure meteorological factor leads to slightly lower pressure and higher windspeed along with the cold ozone depleted conditions of PC3. This high BrO_LTCol event is emblematic of ozone depletions that occur when BrO is more vertically distributed in a low pressure system, enhanced by SSA that increases aerosol extinction and the bromide-containing surface available for reactive bromine recycling (Jones et al., 2009; Luo et al., 2018; Zhao et al., 2017). The character of such vertically distributed events would depend on the strength of contributions from each factor.

Variance in BrO_surf is explained mainly by the chemical factor (PC3) and the cold stable meteorological factor (PC2). PC2 explains a high amount of variance in BrO_surf and minimal variance in BrO_LTCol. The most important aspects of this meteorological factor are cold temperatures and a stable lowest 1,000 m. Temperature inversions are able to contain Br2 emitted from the surface snowpack (Custard et al., 2017) that is ultimately converted to BrO in lowest layers of the atmosphere (Peterson et al., 2015). PC2 and PC3 working concurrently could create surface based BrO events under a stable boundary layer with periodic ozone depletion, similar to previous surface-based observations (Frieß et al., 2004; Koo et al., 2012; Peterson et al., 2015).

We can approximate environmental conditions during an event when PC2 and PC3 work concurrently at comparable strength by adding the loadings from each PC. The resulting high BrO_surf event occurs under environmental conditions with cold temperatures, increased stability in the lowest 1 km, high aerosol extinction and high surface pressure. While PC2 has a positive ozone loading, the low ozone loading in PC3 may cause ozone depletions while these factors are working concurrently at comparable strength. Surface snowpack is highly porous, and reactive bromine can be recycled on snowpack surfaces into interstitial air (Michalowski et al., 2000; Pratt et al., 2013; Thomas et al., 2011). We propose that snowpack surface is the main source of bromide for eventual recycling to BrO for this type of BrO_surf event. Given this context, the high wind loading in PC2 is most likely related to wind pumping enhancing reactive bromine recycling at the surface (Thomas et al., 2011). Enhancement of total bromide containing SSA surface area by high surface windspeeds as mentioned above could also aid recycling of BrO near the surface.

4.7. Reconciling Previous Findings

High BrO events resembling those identified in the previous section have been observed during individual campaigns reported in the literature. With our large data set, we can see and distinguish between two types of high BrO event on statistically significant and physically distinct scales. Both types of event are correlated with increased wind speed in our analysis, which can lead to high BrO_surf due to wind pumping of Br2 from the snowpack (Thomas et al., 2011) or high BrO_LTCol when bromide rich SSA is lofted into the troposphere (Yang et al., 2008). The reactive bromine event under cold stable windy conditions identified here differs...
from the second strong ozone depletion regime defined in Jones et al. (2009) by low wind speeds. The reactive bromine regime first proposed by Jones et al. (2009) occurs in conditions with increased wind speeds and blowing snow. Blowing snow can increase with wind speed, but blowing snow is also affected by snow conditions, wind direction, and wind characteristics (Sturm & Stuefer, 2013). The average wind speed in this analysis is only 5.6 m/s, below the expected threshold wind speed for SSA formation from blowing snow (6.975 m/s) (Déry & Yau, 2001; Yang et al., 2008). Previous analysis of observations at Utqiagvik did not observe an obvious relationship between BrOLTcol and windspeed (Peterson et al., 2015). An earlier analysis of O-Buoy ODEs observed across a range of temperatures below freezing found low windspeeds (median of 3.6 m/s) during ODEs (Halfacre et al., 2014). Previous ground-based studies used BrO observations of both types of reactive bromine event from early O-Buoys and Utqiagvik but did not always separate between high BrOLTcol events and high BrOsurf events as we have done here.

Ground-based measurements may be more likely to detect surface-based reactive bromine events and space-based observations may be more likely to detect reactive bromine events in low-pressure systems. The BROMEX field campaign observed different types of high BrO events, with shallow events similar to the high BrOsurf event identified in the previous section and events associated with high extinction similar to the high BrOLTcol event in the previous section (Simpson et al., 2017). Satellites have also observed Arctic low-pressure systems enabling SSA formation and subsequent reactive bromine production and ozone depletion (Zhao et al., 2017). We find that both types of event can regularly occur during the Arctic springtime and can be distinguished by differing meteorological conditions and vertical distribution of BrO. Further ground- and space-based observations may be used in harmony to gain a better understanding of reactive bromine events.

### 4.8. Prediction of BrO From Three Principal Components

Predictive modeling allows us to test the skill of the factors identified above in predicting BrO. The model generated by PCR predicts reactive bromine vertical column densities across 5 years and vast regions of the Arctic within observational uncertainty nearly 50% of the time. Figure 4 shows predictions clustered near the 1:1 line with observations; however, the highest observed values are much larger than the largest predictions. Replicating the upper ranges of observed BrO may be difficult due to the abrupt and nonlinear processes inherent in bromine explosion chemical mechanisms. Our linear model does not attempt prediction when ozone is below 2 nmol/mol. This leads to issues with modeling the extended high BrO event in late May 2014 (see Figure 5) when ozone falls below 2 nmol/mol and the model has no predictions.

The correlation coefficients between BrO observations and predictions ($R = 0.55$ for BrOsurf and 0.43 for BrOLTcol) are appreciable given that our model attempts to predict observed BrO across many different locations at different dates in the spring across 6 years. The model never predicts BrOLTcol above $3 \times 10^{13}$ molecules/cm$^2$ or BrOsurf above $1.3 \times 10^{13}$ molecules/cm$^2$, which are both less than a third of their respective BrO maxima. Our statistical model can accurately predict when BrO is higher than average but is not able to predict extremely high BrO observations. The skill of this linear model, using almost entirely meteorological variables, shows the importance of including meteorology in any reactive bromine modeling study.

### 5. Conclusion

We examined reactive bromine in a 5-year data set gathered over the Arctic Ocean, as well as at the coastal location of Utqiagvik. We found significant mean BrO varies by a factor of 2–3 between years and is statistically significant. Regional differences are also a factor of 2 with the lowest values in the Beaufort Gyre, but their statistical significance does not reach accepted confidence levels. Seasonally averaged BrO was negatively correlated with the fall minimum sea ice extent in the previous year. From principal component analysis, we identified one chemical factor and two meteorological factors associated with high BrO events.

The chemical factor (PC3) consists of cold, low ozone conditions, and both high BrOLTcol and high BrOsurf are associated with this factor. This chemical factor may act at the same time as the meteorological factors to produce two different types of high BrO events. The first meteorological factor (PC1) consists of meteorological conditions with, low pressure, high wind, thicker mixed layers and decreased stability, which is associated with higher BrO distributed through lower troposphere. When working concurrently with the
chemical factor, this statistical relationship predicts high BrO events that are consistent with previous space-based observations of high BrO in low-pressure systems (Jones et al., 2009; Zhao et al., 2017) and ground-based observations of high BrO during periods of high aerosol extinction (Pöhler et al., 2010). The second meteorological factor (PC2) consists of cold temperatures and increased stability and is associated with increased BrO in the lowest 200 m of the atmosphere. When working concurrently with the chemical factor, this statistical relationship predicts high BrO events that are consistent with previous ground-based observations of surface-based reactive bromine events (Peterson et al., 2015) and field (Pratt et al., 2013) and modeling (Lehrer et al., 2004) studies.

These statistical relationships can predict BrO from local chemical and meteorological variables. The statistical prediction has reasonable skill (R ≈ 0.5) across many sites, regions, and years and is able to predict BrO within observational uncertainty nearly 50% of the time. While the linear model predicts many enhanced BrO events, it underestimates the highest observed BrO values, potentially due to nonlinear chemical or physical processes.

Distinct meteorological factors were consistent with space- and ground-based observations of reactive bromine events. Overall, the data set and model reconcile past literature on Arctic BrO events by providing statistical evidence for two different types of BrO event.

References


