Bromine activation in the high Arctic: four-year time series of BrO columns from Eureka, Canada

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1 Abstract

Bromine explosions and corresponding ozone depletion events are common in the Arctic spring. The sources of reactive bromine are the snowpack and aerosols, but the exact conditions and mechanisms required for bromine release are not well understood. While direct observations of bromine chemistry are challenging, long time series of bromine monoxide (BrO) provide useful insight into the underlying processes of bromine activation. Here we present a four-year time series (2015-2018) of springtime BrO columns retrieved from Multi-AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements from Eureka, Nunavut, Canada (80.1° N, 86.4° W). BrO results are compared to wind observations and surface ozone concentrations to investigate the role of local meteorology. We find that wind direction influences the statistics of BrO observations. Comparisons to in-situ aerosol measurements indicate that high BrO concentrations correlate well with high supermicron aerosol load (likely locally produced sea salt aerosol). BrO columns also show good agreement with columns from the pTOMCAT global 3-D chemistry transport model.

2 Bromine activation

Reactive bromine is released into the atmosphere through an auto-catalytic, multi-phase reaction cycle. BrO, detectable using remote sensing, is a useful indicator of bromine explosions while ozone is still available. Bromine might be released from saline **snowpack** (e.g. Pratt et al., 2013; Custard et al., 2017) or **aerosol particles** (e.g. Frieß et al., 2011; Peterson et al., 2017). Snowpack-related events are commonly associated with low winds and a shallow, stable boundary layer, while strong winds and high aerosol extinction might favor bromine release and recycling from aerosols. The impact of high winds and blowing snow is still uncertain. Blowing snow might act as a direct bromine source, or as a source of sea salt aerosol (**SSA**) through sublimation of lofted, saline snow particles (e.g. Zhao et al., 2017). The resulting SSA particles are likely to be large (>1 µm) and therefore short-lived.

This poster examines the roles of wind speed and aerosol particles in bromine activation at Eureka.

3 Datasets

BrO measurements presented here were made using Multi-AXis Differential Optical Absorption Spectroscopy (**MAX-DOAS**, Figure 1). Low elevation angle measurements of scattered light have longer tropospheric path lengths. Taking the ratio of these low elevation angle measurements and a zenith-sky spectrum yields good sensitivity to near-surface absorbers. The result of the retrieval is the differential Slant Column Density (dSCD).

BrO profiles were retrieved from dSCDs using a two-step optimal estimation approach (Frieß et al., 2011). First, aerosol extinction profiles were retrieved from oxygen collision complex (O_4) dSCDs. The extinction profile then served as a forward model parameter for the BrO retrieval. Profiles were accepted only if they had degrees of freedom for signal of 0.7 or higher, with aerosol extinction below 5.

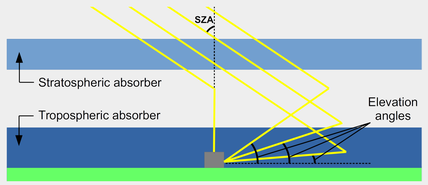

Figure 1: MAX-DOAS viewing geometry. SZA is the solar zenith angle. Elevation angles of -1°, 1°, 2°, 5°, 10°, 15° and 30° were used in this study.

The instrument used here is the PEARL Ground-Based Spectrometer (PEARL-GBS), a triple grating spectrometer with a cooled CCD detector and a suntracker system. MAX-DOAS measurements were taken in the UV, for solar elevations above 4°.

In-situ aerosol measurements were made using an optical particle counter (**OPC**) that measures particles with diameters between 0.3 and 20 microns in 6 size bins. Surface ozone was measured using a Thermo Fisher Scientific Model 49i **ozone analyser**. Ozonesondes and radiosondes were used in the BrO retrieval.

The **model BrO** dataset was taken from the pTOMCAT global 3-D chemistry transport model (2.8° x 2.8°, with 31 vertical layers and 1 hour time resolution; Yang et al., 2005). Bromine sources in the model include SSA from open ocean and from blowing snow, but the model contains no snowpack chemistry, meaning that large BrO enhancements are due to SSA-sourced bromine.

Figure 2: Location of Eureka (Ellesmere island, Nunavut) in the Canadian high Arctic, at 80.1° N, 86.4° W.

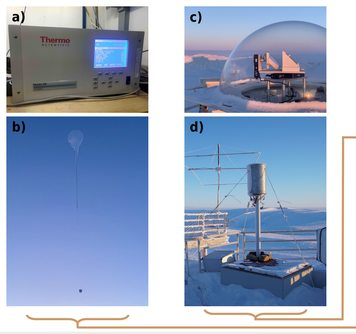

Figure 3: **a)** TFS 49i ozone analyser, **b)** ozonesonde just after launch, **c)** the MAX-DOAS input optics, **d)** aerosol inlet for the OPC.

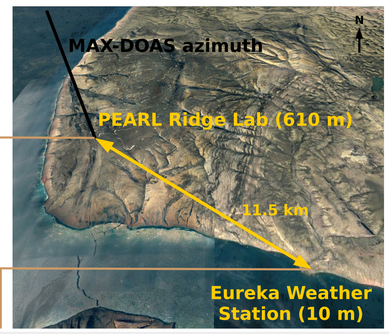

Figure 4: Map of Eureka, with the Polar Environment Atmospheric Research Laboratory (PEARL) and the Weather Station.

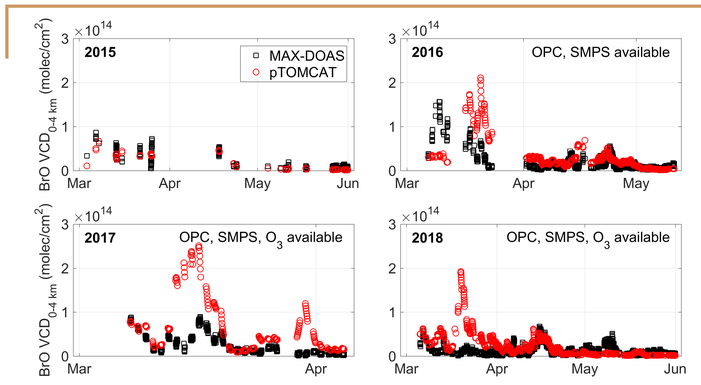

Figure 5: Yearly time series of 0-4 km BrO columns retrieved from MAX-DOAS measurements, along with modeled data from pTOMCAT interpolated to the measurement site. The model generally captures the bromine events, but often overestimates the BrO concentrations.

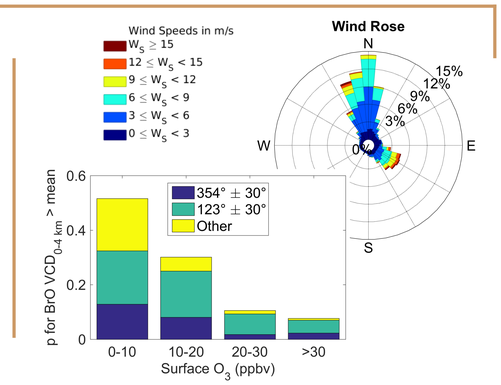

Figure 6: Bar plot of BrO columns above the overall mean column, grouped by surface ozone concentrations and wind direction. The wind rose shows March-May winds for PEARL.

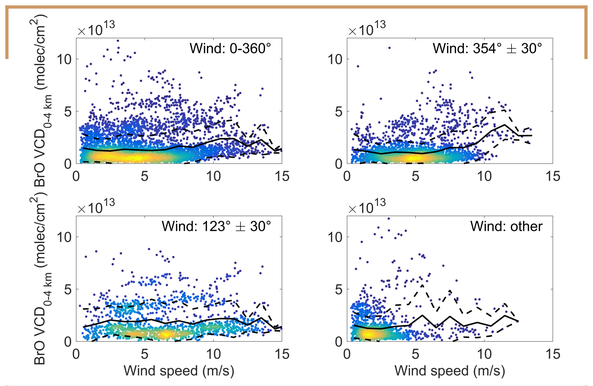

Figure 7: Normalized density plots of BrO columns vs. wind speed, for various wind directions. The black lines are the mean and standard deviation using 1 m/s bins. There is an increase of mean BrO columns for strong northerly winds only.

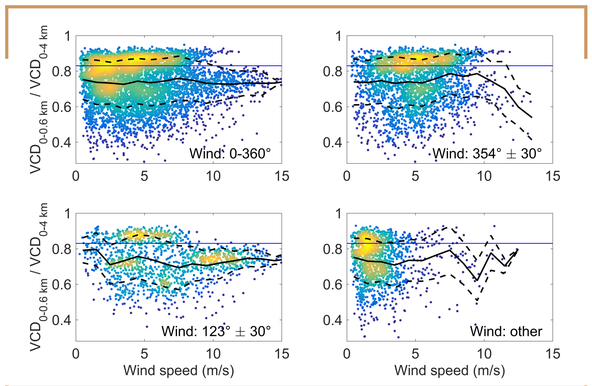

Figure 8: As Figure 7, with ratios of 0-0.6 km and 0-4 km BrO columns. The blue lines represent the a priori ratio. Near-surface enhancement is typical for low wind speeds, while high winds seem to generate vertically distributed events.

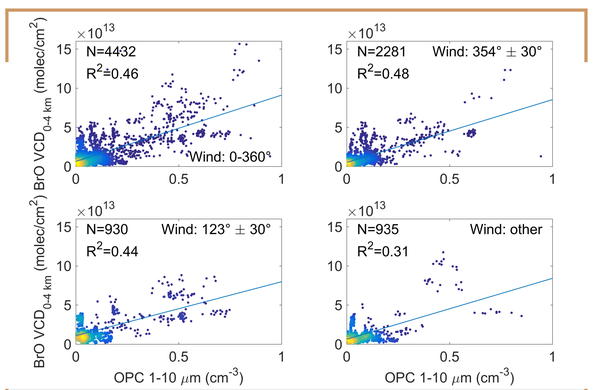

Figure 9: Correlation of 0-4 km BrO columns with total supermicron particle concentrations from the OPC, as a function of wind speed. The blue line represents a linear least squares fit to the data.

4 Conclusions

- The pTOMCAT results (with no snowpack chemistry) generally reproduce the observed BrO time series.
  - Model overestimation likely due to representation error and model boundary layer dynamics.
- Bromine explosion characteristics at Eureka depend on wind direction.
  - BrO columns increase with strong northerly winds.
  - Southeasterly winds generate vertically well-mixed bromine explosions, often with less ozone depletion.
- High supermicron aerosol load appears to be a necessary condition for high BrO columns.
  - Accounts for almost half of the BrO variability.
- Separating the roles of transported versus locally generated BrO and aerosols requires more work.

Acknowledgments

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