Measurements of tropospheric bromine monoxide over four halogen activation seasons in the Canadian high Arctic*

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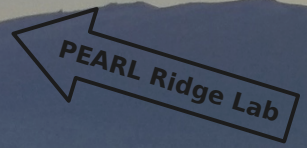

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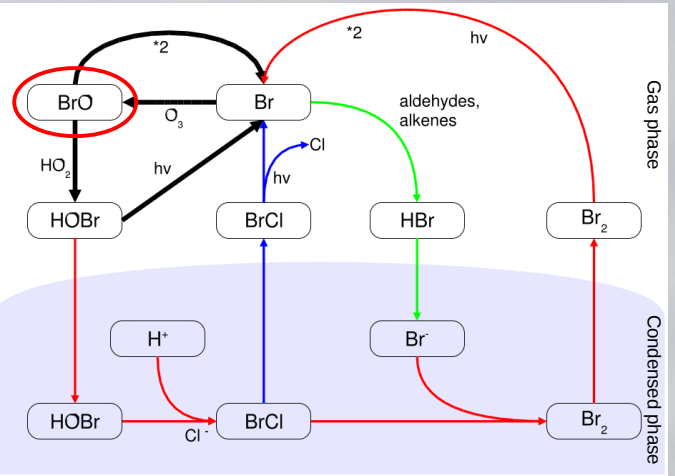
Introduction

- Ozone depletion events (ODEs) are common in the Arctic spring
  - Ozone at near-zero concentrations for hours/days (from ~30 ppbv background)
  - Caused by reactive halogens, terminated by mixing
- Bromine explosions: heterogeneous reaction cycle
  - Autocatalytic; can cause sudden buildup of atmospheric bromine
  - BrO (detectable using remote sensing) is present while ozone is still available
- Bromine sources
  - Snowpack on land and on sea ice [2,3]: calm conditions, shallow ODEs
  - Blowing snow, sea salt aerosols (SSA) likely also contribute
    - Strong winds → well-mixed BrO → aerosols required for bromine recycling aloft [4]
    - Blowing snow is a major source of SSA in the polar winter and spring [5,6]
    - Bromine release from SSA observed in Antarctica [6]
- Ground-based and satellite studies often disagree on dominant source
  - Implicate snow and SSA, respectively, due to different vertical sensitivity
- We present a 4 year dataset of springtime BrO partial columns
  - Ground-based site with sensitivity to the free troposphere

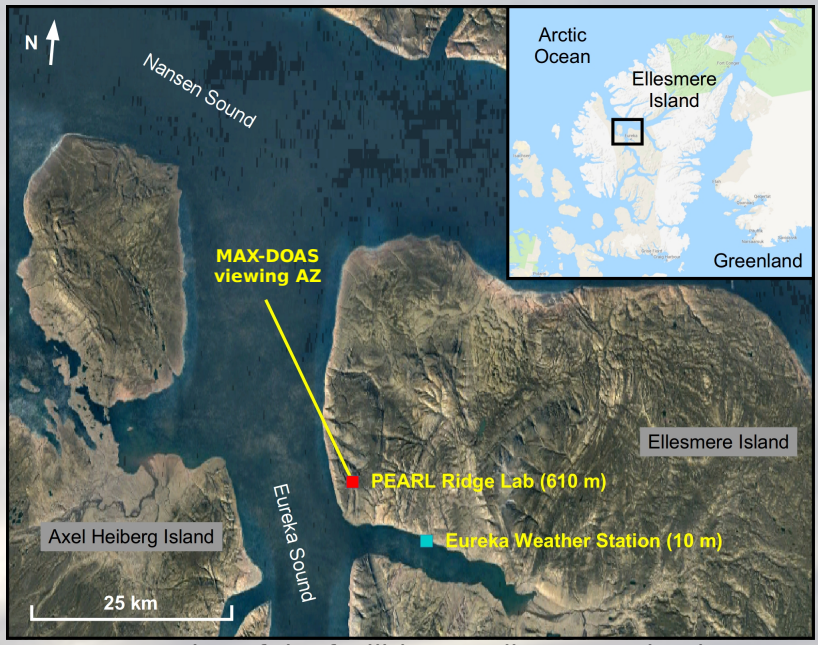


Simplified set of bromine explosion reactions [1]

Blowing snow over the Canadian Arctic

Site description and methods

- Eureka, Canada (80° N, 86° W)
  - Eureka Weather Station (EWS, 10 m): sonde launches, surface ozone
  - PEARL Ridge Lab (610 m): MAX-DOAS, in situ aerosols
- Strong temperature inversions in the spring
  - Stable atmosphere, except for storms (well-mixed BL)
- Bimodal winds: all data categorized by wind direction
  - Northerly (N): 354° ± 30°, ~50% of observations
  - Southeasterly (SE): 123° ± 30°, ~25% of observations
- AOD, BrO retrieved using the HEIPRO algorithm [7]
  - 0-4 km, using partial columns only
  - Not sensitive to the surface (sea level)
    - Instrument is at 610 m, lowest elevation angle is -1°
- Given the stable atmosphere, the partial columns often represent BrO above the boundary layer
  - Effect of mountainous terrain not known

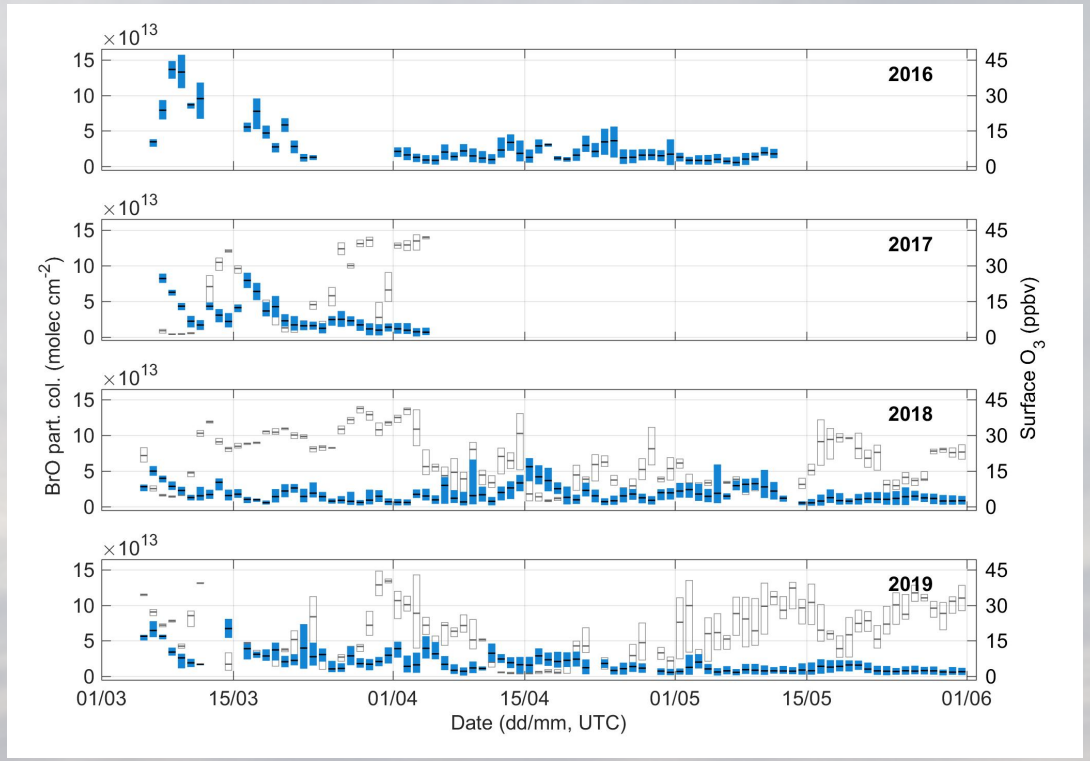


Location of the facilities on Ellesmere Island

The EWS and the PEARL Ridge Lab

Dataset overview

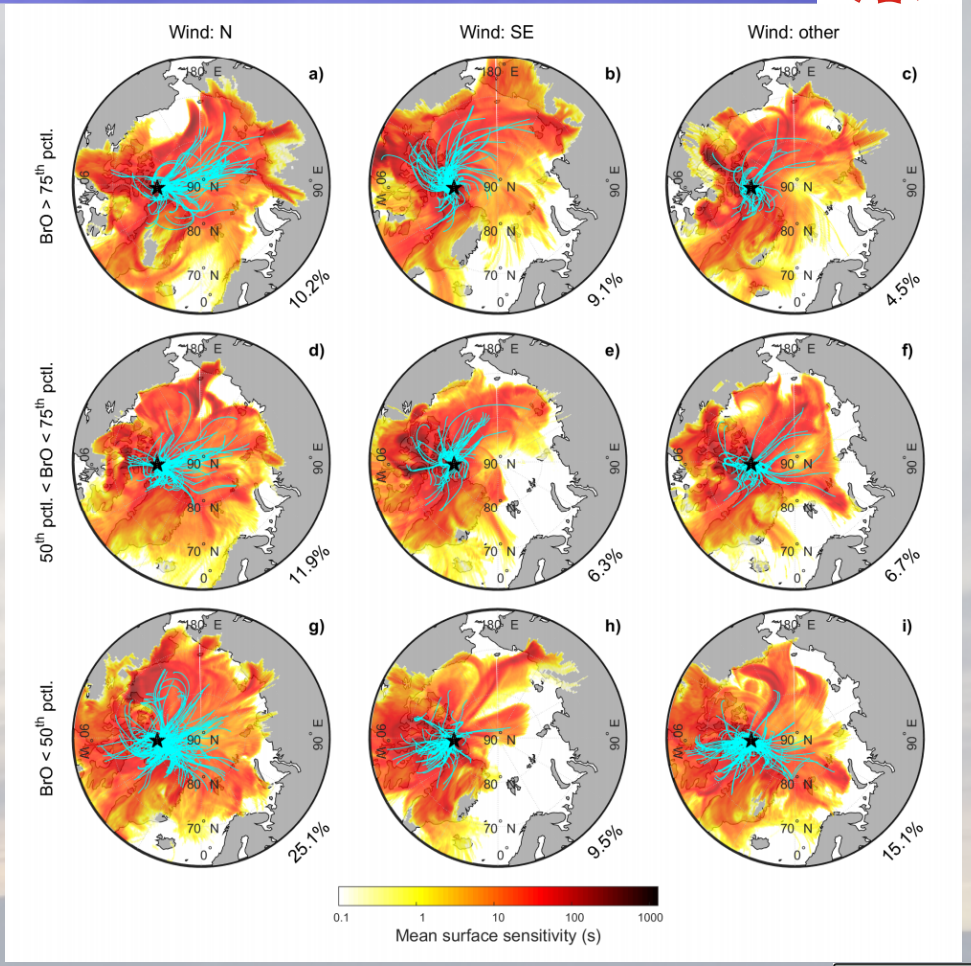
- March – May data for 2016 – 2019
  - Data prior to 2016 not used due to restricted elevation angle range (> 5°)
  - Missing days are whiteout conditions or instrument downtime
- Largest BrO partial columns in March, some activity even in May
  - Unusual pattern in 2018, with low BrO in March, and larger enhancements in April and May
- Close to 50% of surface ozone measurements show depletion (< 15 ppbv)
  - No clear relationship to BrO
    - Includes ozone-depleted air masses with no BrO
  - On average, ozone-depleted airmasses correspond to higher BrO than non-ODE samples



Daily range of retrieved 0-4 km BrO partial columns (blue rectangles) and surface ozone values (empty rectangles) for March to May 2016-2019. The rectangles represent the full daily range of the values, and the black/grey lines show the mean values for each day. Note that the surface ozone instrument was installed in August 2016.

Airmass history varies with local wind direction

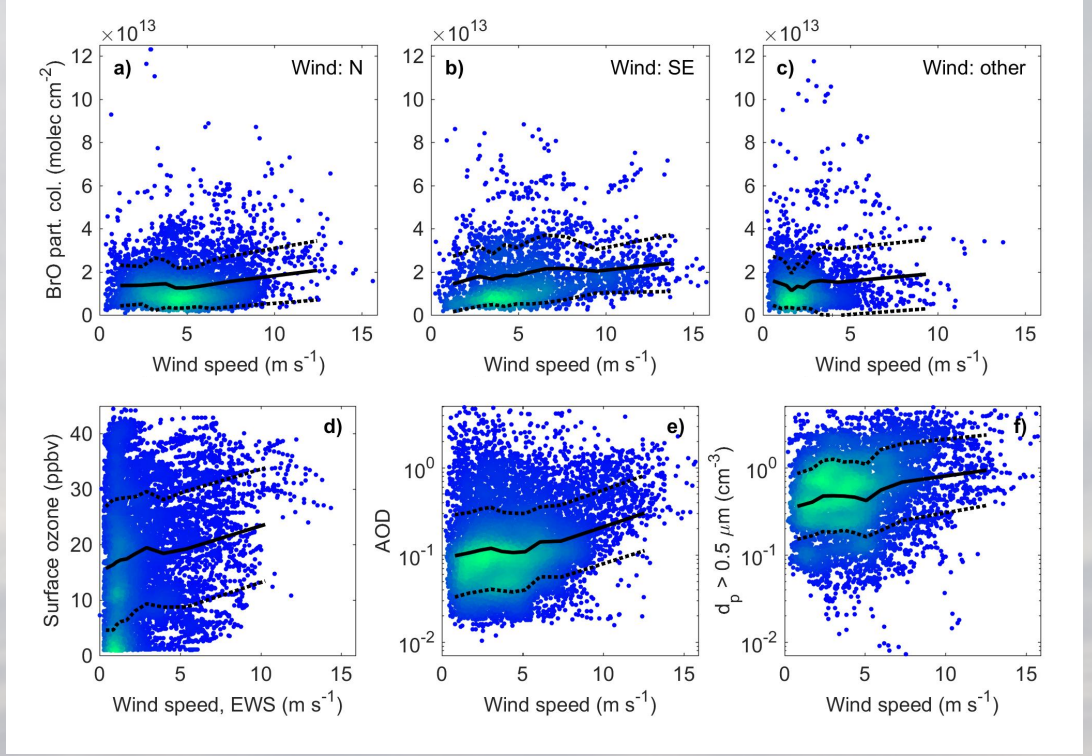
- 3-day FLEXPART back trajectories (approx. every 3h)
- BrO enhancements correspond to distinct airmass histories for each wind direction
  - Moderate and low BrO events differentiate less and less
- N winds and high BrO: air from the Arctic Ocean
  - Sensitivity to Central Arctic
  - Sea ice contact important (slide 7)
- SE winds and high BrO: storms
  - Originating in Canadian, Western Arctic
  - Almost all storm-like trajectories correspond to enhanced BrO
  - Aerosols play larger role (slide 8)



Mean surface sensitivities and individual trajectories for subsets of the FLEXPART back trajectory dataset. The black star indicates Eureka. The rows are grouped by the mean BrO corresponding to each FLEXPART run, and the columns are grouped by mean wind direction. The percentages show the portion of back trajectories that fall into each subcategory (excluding back trajectories with missing wind data)

Local weather plays an important role

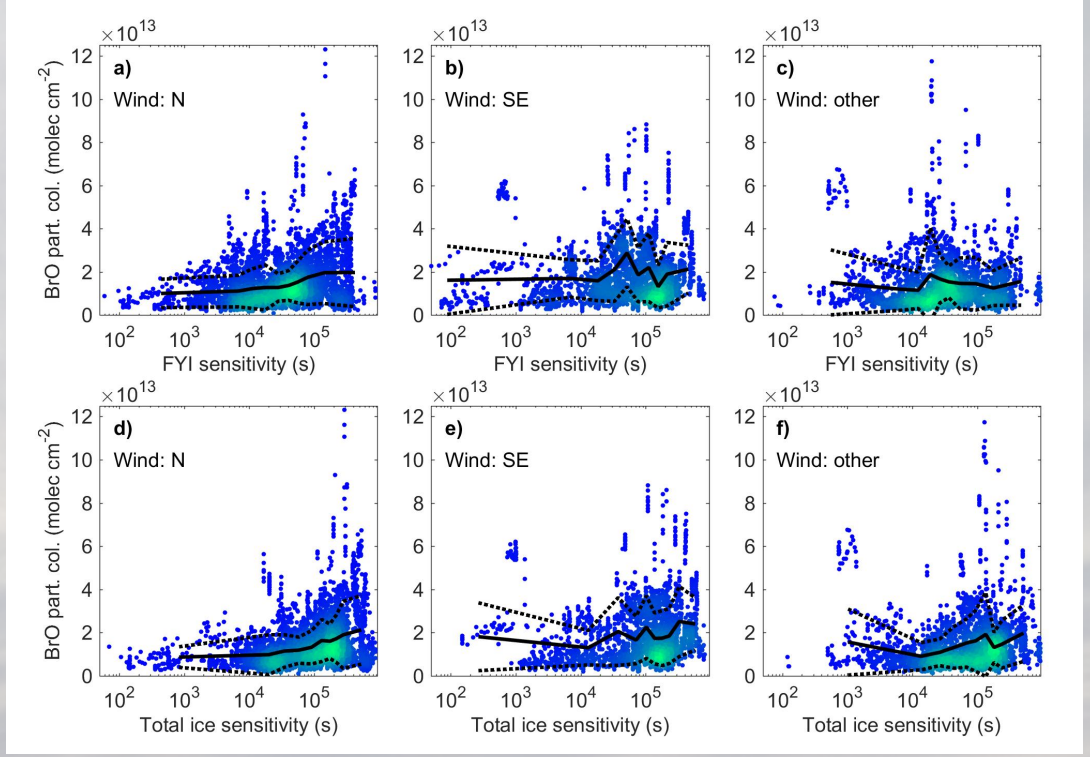
- BrO appears largely independent of 0 to 600 m inversion strength
  - BrO aloft is often decoupled from the surface
  - Missing shallow ODEs that are evident in surface ozone data (weak winds, strong inversion)
- Weak relationship between all BrO and winds
- Strong winds generate vertical mixing
  - Minimum BrO values increase, likely due to mixing
  - At the same time, ODE frequency is reduced
    - Ozone is replenished faster than it is destroyed
    - During storms, ozone generally increases with BrO
  - AOD, aerosol concentrations both increase
    - Blowing snow → local SSA production?
- Maximum BrO values increase with decreasing temperature
  - Temperature explains ~30% of BrO variability
  - BrO enhancements up to 0 °C, highest below -10 °C



0-4 km BrO partial columns as a function of wind speed (a-c), and surface ozone, retrieved AOD, and coarse mode aerosol number concentrations as a function of wind speed (d-f). The color scaling shows the normalized density of the points. The solid lines show the mean for each decile of the data, and the dotted lines indicate the corresponding one standard deviation.

FYI contact is important for N winds only

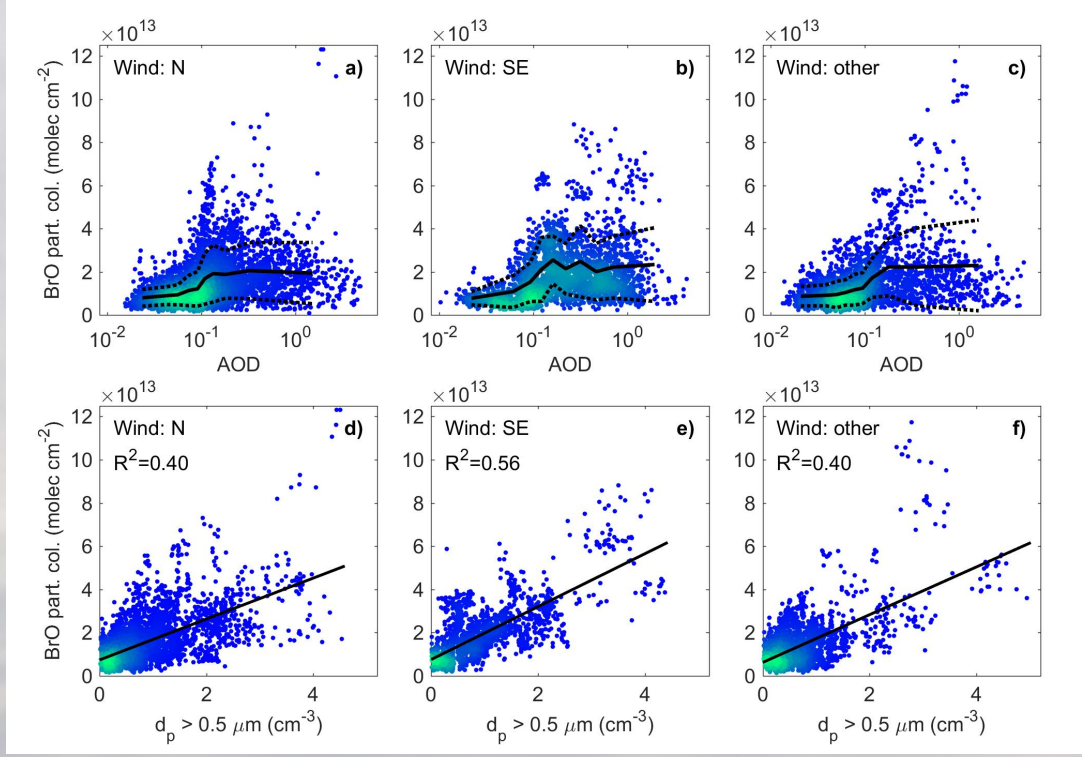
- Using the EASE-Grid sea ice age dataset
  - Ice sensitivity calculated for each FLEXPART run
- All wind directions show similar sensitivity to first year sea ice (FYI)
  - Mean BrO values increase for N winds only
    - Air from over the Arctic Ocean
- FYI+MYI is a slightly better predictor of BrO
  - Eureka is close to the pack of perennial ice
  - Increased correlation (compared to FYI only) noted before [8]
- Atmospheric processes could contribute to bromine release from MYI
  - Deposition of SSA, acidification by Arctic haze
  - Dataset contains several BrO events with fraction of MYI sensitivity greater than 0.9
  - Bromide depletion observed in snow on MYI [9]



0-4 km BrO partial columns as a function of (a-c) FYI sensitivity and (d-f) all sea ice sensitivity, calculated using FLEXPART sensitivity maps and EASE-Grid sea ice age. The color scaling shows the normalized density of the points. The solid lines show the mean BrO partial column for each decile of the sensitivity data, and the dotted lines indicate the corresponding one standard deviation.

Coarse mode aerosols are necessary and sufficient

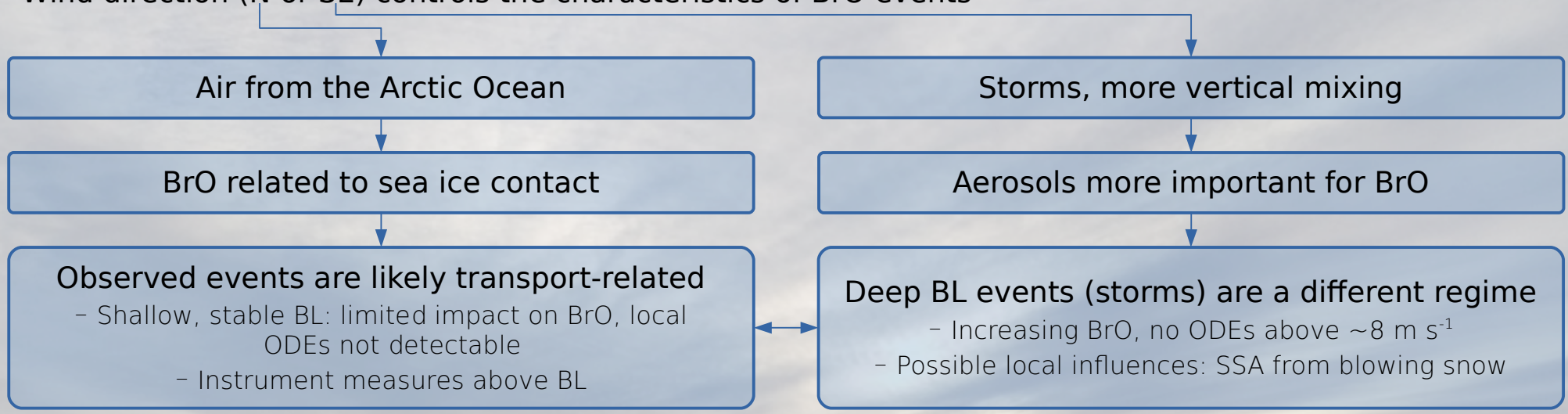
- BrO values increase with AOD
  - Larger portion of high AOD data for SE (storms)
  - Not sufficient for high BrO [4]
- In situ aerosol data from the PEARL Ridge Lab
  - Accumulation mode ($0.1 < d_p < 0.5$ µm)
  - Coarse mode ($d_p > 0.5$ µm)
- Good correlation with coarse mode aerosols
  - Best for SE winds
  - Highest BrO values are only observed when aerosol concentrations are also high
  - No correlation with accumulation mode aerosols
    - Arctic haze, no direct role in bromine release
- Coarse mode aerosols are likely SSA from blowing snow [6]
  - No other sources, no open water along trajectories
  - Data consistent with local SSA production (slide 6)



0-4 km BrO partial columns as a function of (a-c) retrieved AOD, and (d-f) coarse mode aerosol number concentrations. For panels a-c, the solid lines show the mean BrO for each decile of the data, and the dotted lines indicate the corresponding one standard deviation. For panels d-f, the solid lines show a linear least squares fit, with the R^2 values indicated on the plots.

Summary

- BrO above the boundary layer is common in Eureka from March to May
- Wind direction (N or SE) controls the characteristics of BrO events



- MYI might also contribute to bromine release, after salting/acidification through atmospheric processes
- Aerosols are necessary to maintain BrO (for all wind directions), since we often measure above the BL
  - Coarse mode aerosols (likely SSA) are necessary and sufficient to observe high BrO at Eureka
- The impacts of topography on the BL need to be quantified to understand the vertical distribution of BrO over mountainous terrain (much of the Canadian Arctic)

References and contact

- Contact: Kristof Bognar

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  - Canadian Arctic ACE/OSIRIS Validation Campaigns (Kaley Walker): CSA, ECCC, NSERC, NSTP
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- References

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