Evaluating sources and concentrations of reactive bromine in the Arctic using ground observations and GEOS-Chem

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Remote sensing of reactive bromine

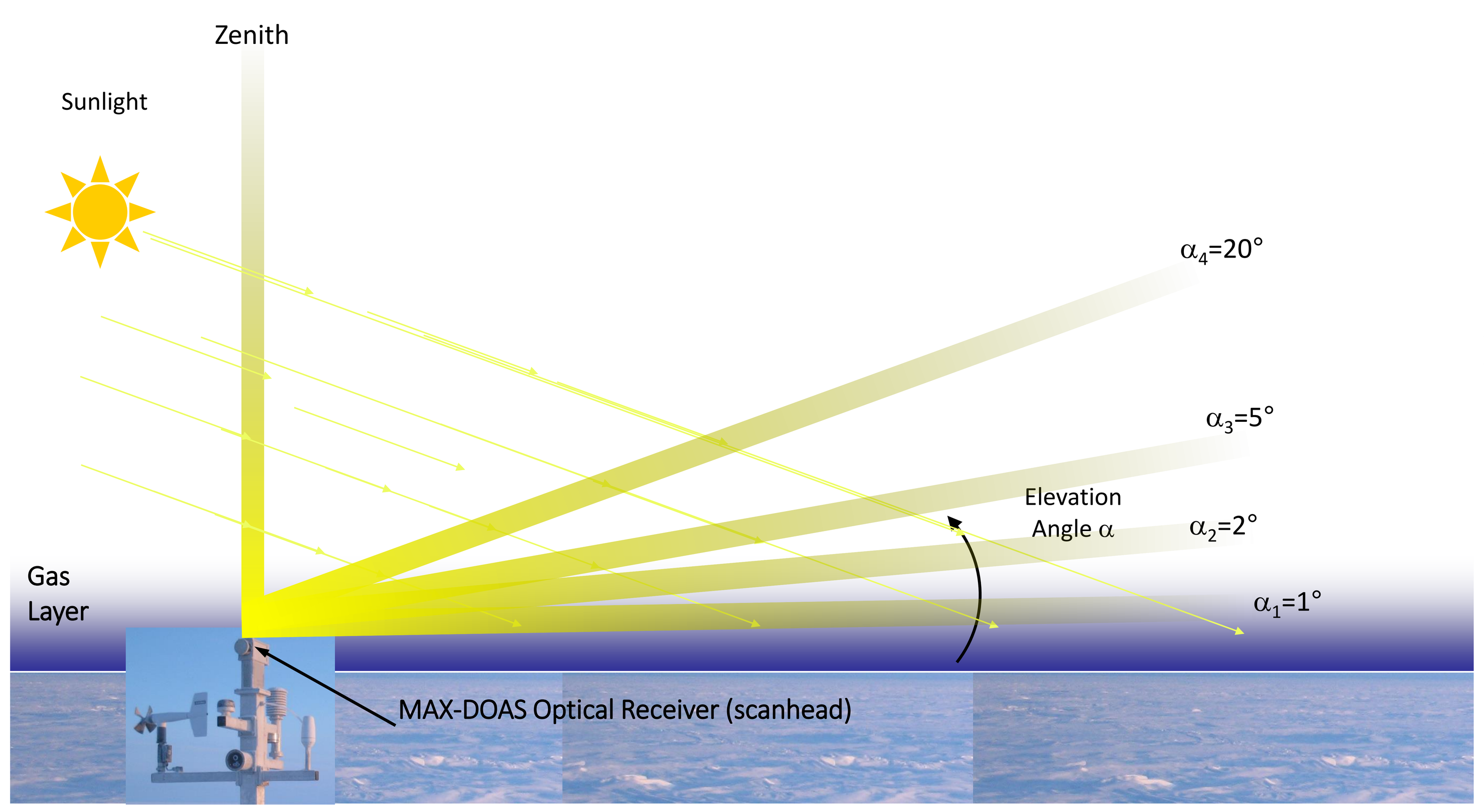

Figure 1: Illustration of MAX-DOAS view angles as deployed aboard the OBUOY. Adapted from Hönninger et al.,2004

- Reactive bromine is produced seasonally in the Arctic spring, and can lead to ozone depletion and mercury oxidation
- Spectrometers measure the vertical column density of a gas along a path (molecules/cm^2), which is an integral of the concentration of the gas along the entire path length (slant column density, shown in yellow).
- Multi-axis differential optical absorption spectroscopy (MAX-DOAS) shown in Figure 1 scans for the reactive bromine species bromine monoxide (BrO)
- Optimal estimation based inversion of all slant column densities retrieved:
  - **BrO_{VCD200}** the integrated vertical column density of BrO from the surface to 200 m
  - **BrO_{LTVCD}** :the integrated lower tropospheric vertical column density of BrO from the surface to 2000 m (Peterson et al, 2015).
- Increased BrO has been observed under storm conditions with high wind and aerosol (Frieβ et al., 2011) and under inversions in the lowest layers of the atmosphere (Peterson et al., 2015)
- Surface-based remote sensing observations can test model skill
- We want to determine which conditions are most important in driving BrO increase
- We want to test the ability of GEOS-Chem to replicate springtime BrO increases

Two model approaches

Dynamical: GEOS-Chem with blowing snow sea salt aerosol particle (SSA) production (Huang et al., 2018) in GEOS-Chem

Statistical: PCA of O-Buoy dataset to determine meteorological controls on BrO, then predict BrO using MERRA-2 and ozone

Compare each model to O-Buoy BrO observations (Fig 3)

GEOS-Chem modeling

- GEOS-Chem version 12.1.0, Tropospheric Chemistry Scheme (Parrella et al, 2012)
- MERRA-2, 4° x 5° Resolution, 47 vertical layers
- Blowing snow Br_y from Huang and Jaeglé (2016), including seasonal cycle of snow salinity (Huang and Jaegle, 2018)
- **Implemented new diagnostic to integrate vertical column densities along O-Buoy path using planeflight module:**
  - BrO in lowest 200 m (BrO_{VCD200}) in units of molecules/cm^2
  - BrO in lowest 2000 m (BrO_{LTVCD}) in units of molecules/cm^2
  - BrO in all layers of troposphere in units of molecules/cm^2
  - BrO in all layers of atmosphere, in units of molecules/cm^2
- Integrated vertical column densities in the in the lowest 200 m and 2000 m can be directly compared to MAX-DOAS measured BrO_{VCD200} and BrO_{LTVCD}
- Integrated vertical column densities in the troposphere and in all layers of the atmosphere can be compared to satellite retrievals

Beaufort Gyre Spring 2015, O-Buoy 11

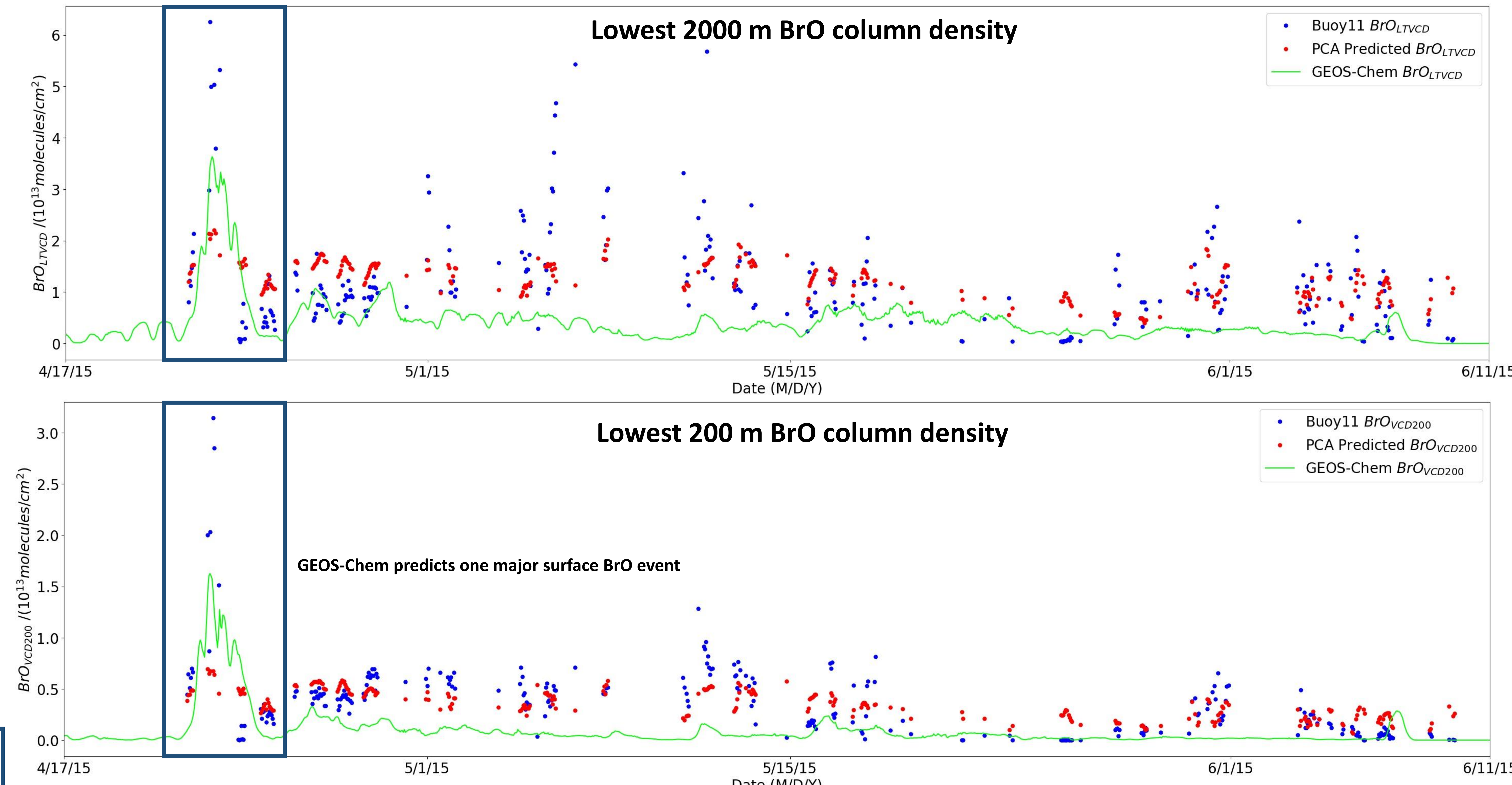

Figure 3: Timeseries comparing modeled and observed BrO in the Beaufort Gyre north of Alaska in Spring 2015 on O-Buoy 11. **Top: BrO_{LTVCD}** or BrO in the lowest 2 km of troposphere. O-Buoy observations shown in blue, PCA predictions shown in red, and modeled GEOS-Chem BrO shown in green. **Bottom: BrO_{VCD200}** or BrO in the lowest 200 m of troposphere. O-Buoy observations shown in blue, PCA predictions shown in red, and modeled GEOS-Chem BrO shown in green.

Principal components analysis modeling

- O-Buoy BrO observations span from 2011-2016 in the Beaufort Gyre and North Pole regions. Added observations at Utqiagvik (Barrow) from 2012-2016
- Combined with meteorological observations to determine meteorological controls on BrO
- **In-Situ Variables: BrO_{LTVCD} BrO_{VCD200} T** : Ambient temperature at 2 m **O3:** Ozone mixing ratio **EXT**: Aerosol extinction at 361 nm
- **MERRA-2 Variables: V_{wind}** : 10 meter windspeed **PBLH**: Richardson boundary layer height **ΔP_{hr1}** : Change in pressure from previous hour **$\Delta\Theta_{1000m}$** : Change in potential temperature from 2m to 1000m **$\Delta\Theta_{100m}$** : Change in potential temperature from 2m to 100m **P** : Surface pressure at 2 m
- Principal component analysis (PCA) run on dataset, found 2 principal components (PC) related to BrO explaining variance greater than unity
- PC Scores calculated using all variables except BrO_{LTVCD} and BrO_{VCD200}. For each other variable in PC, multiplied observation by loading, and added together for score
- PC scores used to predict BrO in Figure 3

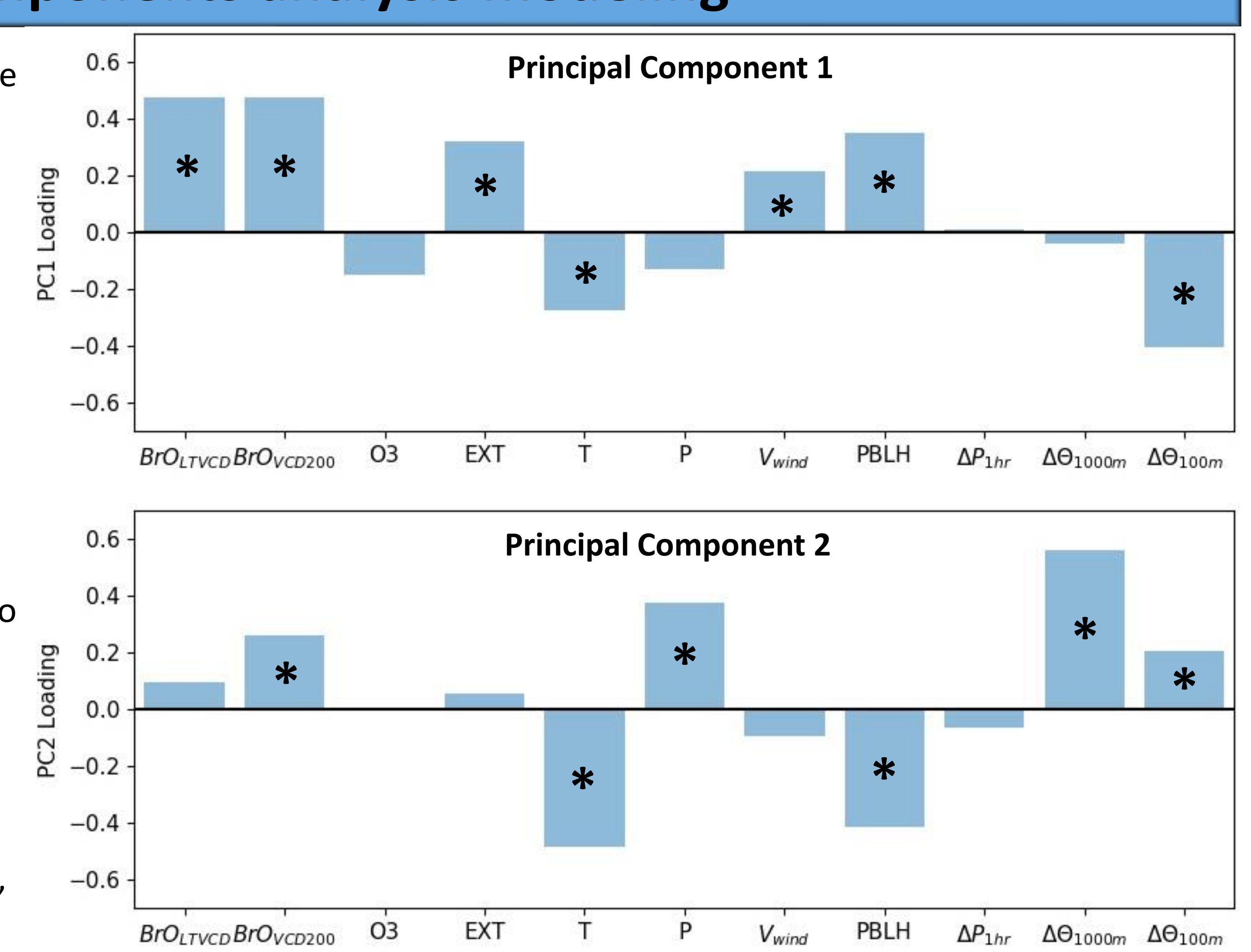

Figure 2: Principal component loadings from PCA. Variables standardized to μ=0, σ = 1. PC1 describes a relationship where an increase of 0.48 σ in Column BrO is correlated to a decrease in ozone of 0.15 σ, and so on for all variables. * denotes loading > 0.2.

Main takeaways

- Integrated vertical column density : useful diagnostic for model comparison to ground-based remote sensing
- Statistical and dynamical models underestimate observed peak BrO
- GEOS-Chem with blowing snow simulates major BrO event
- Statistical model suggests importance of cold, shallow inversion conditions to column BrO in lowest 200 m

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Mille grazie a mia famiglia e i miei amicci!