

OMI Measurements of Bromine Monoxide and Implications for Missing Sources of Polar Bromine in GEOS-Chem

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Background

- Bromine Monoxide (BrO) is the most frequently measured inorganic bromine compound
 - Measured by the Ozone Monitoring Instrument (OMI), GOME-2, and TROPOMI
 - Stratosphere is a large fraction of column measurements
- Tropospheric bromine:
 - Lowers ozone, alters NO_x and HO_x cycles, and increases Hg bio-uptake
 - Marine and polar sources (algae and sea salt)
 - Polar sources not included in GEOS-Chem





Research Goals



- Polar spring "bromine explosion" events
 - Not included in GEOS-Chem model
 - Associated with near complete removal of surface ozone
 - Detectable by OMI (e.g., Choi et al., 2018)
 - Potential mechanisms: blowing snow, first year sea ice, snowpack
- Development of a simplified polar climatology
 - Identify regions of elevated bromine using OMI BrO
 - Requires accurate estimate of stratosphere
 - Towards assimilation in GMAO GEOS-Composition Forecast system

OMI Total BrO 20080304





GEOS-Chem Stratospheric Bromine Monoxide (BrO)

- Simulated for 2007 2010
 - MERRA-2 reanalysis meteorology
 - GMI stratospheric chemical mechanism
 - Sea-salt debromination turned off
- Stratospheric BrO sensitive to reactions with O_3 and NO_2
 - Microwave Limb Sounder (MLS) retrieves profiles of O_3
 - OMI stratospheric NO₂ calculated using a climatology







GEOS-Chem Stratospheric Bromine Monoxide (BrO)

- Simulated for 2007 2010
 - MERRA-2 reanalysis meteorology
 - GMI stratospheric chemical mechanism
 - Sea-salt debromination turned off
- GEOS-Chem total column BrO biased low
 - Stratospheric loading of bromine in close agreement with WMO Ozone Assessment recommendation (~ 20 ppt)
 - OMI total column adjusted for ~0.5 ppt tropospheric BrO (Brockway et al., in prep)









Global Total Column BrO



- GEOS-Chem biased low compared to OMI
 - 50S to 50N: 1.2 ± 0.2 × 10¹³ molec cm⁻²
 - Consistent with GOME BrO global model comparisons (Falk & Sinnhuber et al., 2018; Fernandez et al., 2019)

Potential sources of bias:

- 1. Free troposphere
 - Heterogeneous recycling, washout rates
- 2. Stratosphere
 - BrONO₂ reaction kinetics
 - Stratospheric bromine loading

Non-polar (50S to 50N) mean bias



Arctic Spring



- Dynamically variable stratosphere in early spring
 - Able to isolate elevated BrO

- Preliminary analysis:
 - Threshold = non-polar mean bias + 3σ
 1.8 × 10¹³ molecules cm⁻²
 - Calculate monthly mean climatology







Arctic Spring

- Large bias in polar spring expected
 - Seen prominently March to May
- Consistent with missing snow/ice source
 - Proposed mechanisms: blowing snow, first year sea ice, snow pack chemistry
 - Can simplified climatology improve monthly mean surface ozone?





Arctic Summer

- Lower bias in Arctic June
 - No significant bias in July and August
- Late spring seasonality
 - Increase in stratospheric NO₂, larger role in BrONO₂ kinetics
 - Investigate impact of:

 $BrO + NO_2 + M \rightarrow BrONO_2 + M$

 $BrONO_2 + HCl \xrightarrow{het} BrCl + HNO_3$



Antarctic Spring



- Seen prominently September to November
- Bias persistent for spring and summer







Antarctic Summer

- Persistent bias in southern ocean
- Likely multiple sources for bias
 - Sea salt aerosol debromination
 - Biogenic short-lived organic bromine



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Summary and Future Directions

- GEOS-Chem stratosphere can:
 - Capture the expected bromine loading and daytime chemistry
 - Identify missing polar tropospheric sources using OMI
- Test ability of climatology to reproduce monthly mean ozone observations
 - Compare to station O₃ and BrO
 - Future: investigate daily BrO flux

