Use of GMI to Study Tropospheric and Stratospheric Bromine Budgets

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Abstract

Use GMI to investigate the importance of short-lived bromine compounds to tropospheric and stratospheric chemistry, including trends in column ozone. We will examine the short-lived gases $CHBr_3$, CH_2Br_2, CH_2BrCl, $CHBr_2Cl$, $CHBrCl_2$, and $C_2H_4Br_2$ by developing emissions inventories and defining removal rates. These short-lived gases will be modeled individually and then all bromine compounds combined. The resulting BrO will be compared with observations from GOME, SCIAMACHY, and in situ instruments.

Motivation

Measurements of stratospheric BrO imply Br_y concentrations that are 4-8 pptv greater than models which include only CH_3Br + halons. [Salawitch et al., 2004]

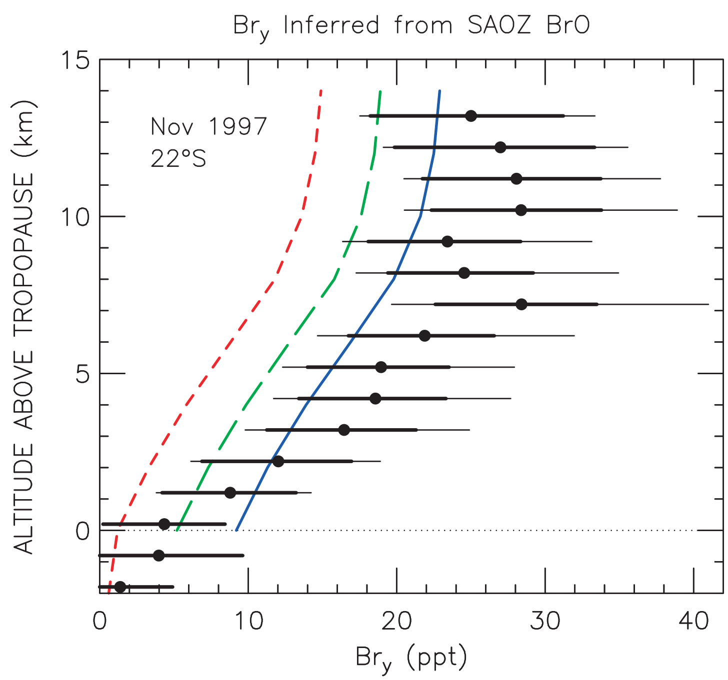

Figure 2 of Salawitch et al. [2004]:

Profiles of Br_y inferred from SOAZ BrO [Pundt et al., 2002] at 22°S in Nov 1997 compared with profiles from the AER 2-D model using Br_y^{TROP} of 0 (red), 4 (green), and 8 (blue) ppt.

Motivation

Sensitivity studies with the AER 2-D model show that an additional 4-8 pptv of Br_y has a large effect on stratospheric ozone trends, particularly in times of enhanced aerosol loading. The additional BrO provides a reaction partner to ClO, leading to more ozone depletion and better agreement between modeled and measured trends [Salawitch et al., 2004]. The GMI strat-trop model provides a unique tool to address the effects of short-lived gases in both troposphere and stratosphere.

Importance to Tropospheric Chemistry

von Glasow et al., [2004] did a 3-D study of tropospheric bromine with simplified emissions and found $Br_y \sim$ 1-6 pptv in troposphere

- BrO/Br_y dependent on het processing of $BrONO_2$ and HOBr (links aerosol to bromine)
- Partitioning important for washout and deposition of HBr, HOBr, and $BrONO_2 \Longrightarrow$ stratospheric Br_y impacted
- tropospheric ozone depletion due to Br_y of up to 15%
- effects on OH, HO_2, and H_2O_2 up to 20%
- DMS+Br, DMS+BrO reactions result in reduced DMS, less SO_2 production

Importance to Stratospheric Chemistry

- Bromine is ∼45 times as effecive as chlorine for ozone removal
- Excess bromine from short-lived substances will impact lower stratosphere most
- Catalytic ozone loss cycles involve BrO+ClO and HOBr+hν
- BrO provides reaction partner for ClO, amplifying effects of anthropogenic chlorine

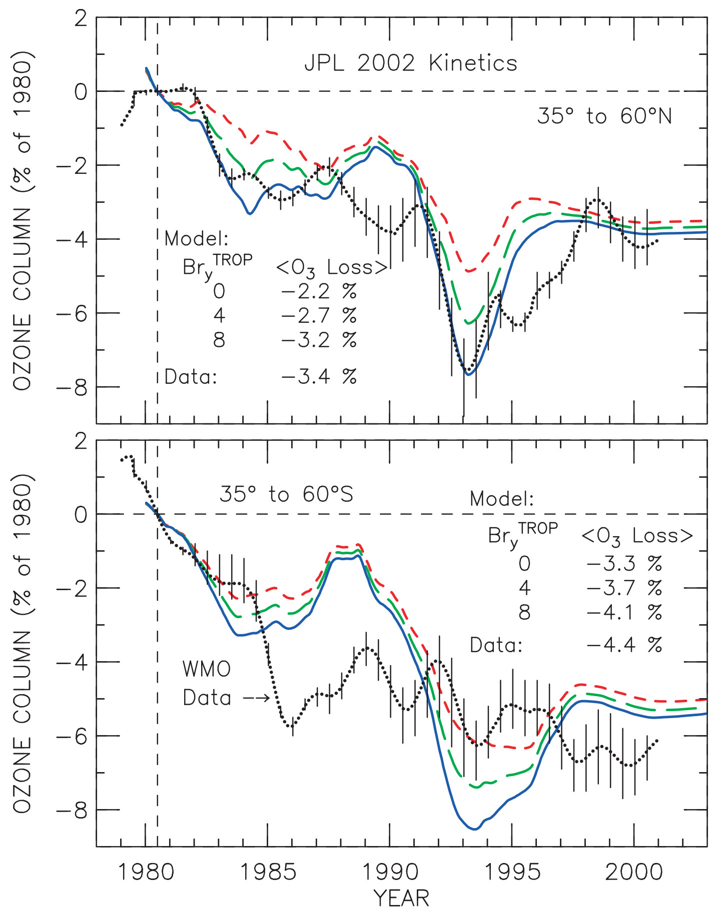

Figure 4 of Salawitch et al. [2004]:

Calculated change in column ozone relative to 1980 levels found using the AER 2-D model for Br_y^{TROP} of 0, 4, and 8 ppt for 35-60°N (top) and 35-60°S (bottom) compared to observed trends [WMO, 2003]. Numerical values represent the average of the modeled and measured ozone depletion from 1980 to 2000.

Relevant Compounds and Sources

compound	abundance	source
CH_3Br	9-10 ppt	fumigation, biomass burning, phytoplankton
$CHBr_3$	0.6-3.0 ppt	phytoplankton, marine macroalgae
CH_2Br_2	0.8-3.4 ppt	phytoplankton, macroalgae
CH_2BrCl	0.1-0.5 ppt	phytoplankton, macroalgae
$CHBr_2Cl$	0.1-0.5 ppt	phytoplankton, macroalgae
$CHBrCl_2$	0.12-0.6 ppt	phytoplankton, macroalgae
$C_2H_4Br_2$	0.3-0.5 ppt	industrial, biogenic?

Relevant Compounds, Emissions, Lifetimes

compound	abundance	emissions	lifetime
CH_3Br	9-10 ppt	112-454 Gg/yr	0.7 yr
$CHBr_3$	0.6-3.0 ppt	200-285 Gg/yr	26 days
CH_2Br_2	0.8-3.4 ppt	55-67 Gg/yr	4 months
CH_2BrCl	0.1-0.5 ppt	2.9 Gg/yr	5 months
$CHBr_2Cl$	0.1-0.5 ppt	4.2-12 Gg/yr	69 days
$CHBrCl_2$	0.12-0.6 ppt	5.5-7.0 Gg/yr	78 days
$C_2H_4Br_2$	0.3-0.5 ppt	77-151 Gg/yr	84 days

Task I: Single Species Calculations

- Run single-species tropospheric GMI on AER computers
- Emission inventories: simple or parameterized by biological productivity of ocean, wind speeds, etc.
- Rates for removal reactions incorporated
- Compare calculated concentrations of individual bromine gases to available observations
- Adjust sources or perform sensitivity studies

Task II: Calculation of Br$_y$

- Incorporate sources and chemical rates of short-lived bromine compounds into GMI (core team)
- Use full strat-trop GMI at GSFC (core team) with all organic bromine compounds to calculated Br$_y$ in trop and strat
- Br$_y$ removal depends on tropospheric partitioning and solubility
- Compare calculated BrO with observations from GOME, SCIAMACHY, and aircraft campaigns. Column BrO data also is available.
- Sensitivity to met fields and OH fields

Task III: Atmospheric Impact Calculations

- Analyze tropospheric changes caused by short-lived bromine compounds, examining O_3, HO_x species, NO_x species, DMS
- Examine interactions between tropospheric bromine and sulfur
- Stratospheric calculations for 1980 and 1993 (high aerosol), with and without short-lived bromine compounds
- Time-dependent hindcast for ozone trend possible