Halocarbons in the TTL: the roles of oceanic emissions and atmospheric transport

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Motivation

Halogen budget $\rightarrow$ Ozone depletion

Stratosphere

Tropical Tropopause Layer (TTL)

Troposphere

Chemical transformation and washout

Very short lived substances

DOM/POM

$hv$

Halocarbons $(\text{CHBr}_3, \text{CH}_2\text{Br}_2, \text{CH}_3\text{I})$

Ozone chemistry

Oxidative capacity

Aerosol formation

Distribution in the TTL?
Method

**Global bottom-up approach using in-situ observations and high-resolution Chemistry-transport modeling**

**Stratosphere**

- Tropical Tropopause Layer (TTL)

**Troposphere**

- Chemical transformation and washout
- Very short lived substances

**Halocarbons**

- \(\text{CHBr}_3\), \(\text{CH}_2\text{Br}_2\), \(\text{CH}_3\text{I}\)

**Comparison to aircraft campaigns**

- Chemical decay: \(\text{OH} \) chemistry or prescribed lifetime
- \(\text{Br}_y\) partition from \(\text{pTOMCAT}\) (Yang et al., 2010)

**Monthly mean emissions derived from observations**

- \(\text{FLEXPART}\) (Stohl et al., 2005)
- \(\text{ERA-Interim}\)

**Method**

Global bottom-up approach using in-situ observations and high-resolution Chemistry-transport modeling
Input for FLEXPART: global VSLS emissions (Ziska climatology)

CHBr$_3$

CH$_2$Br$_2$

CH$_3$I

Ziska et al., ACP, 2013
Results:
Annual mean VMR @ 17 km [ppt]
2008-2010

CH\(_3\)I

CH\(_2\)Br\(_2\)

CHBr\(_3\)
### Results:

Annual mean VMR @ 17 km [ppt]

2008-2010

**CHBr₃**

<table>
<thead>
<tr>
<th>Br [ppt] Based on ...</th>
<th>Inner tropics (10°S-10°N)</th>
<th>Tropics (30°S-30°N)</th>
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</thead>
<tbody>
<tr>
<td>CHBr₃ SG</td>
<td>0.8</td>
<td>0.6</td>
</tr>
<tr>
<td>CHBr₃ SG+PG</td>
<td>1.3</td>
<td>1.1</td>
</tr>
<tr>
<td>CH₂Br₂ SG</td>
<td>2.0</td>
<td>1.7</td>
</tr>
<tr>
<td>CH₂Br₂ SG + PG</td>
<td>2.1</td>
<td>2.0</td>
</tr>
<tr>
<td><strong>All</strong></td>
<td><strong>3.4</strong></td>
<td><strong>3.1</strong></td>
</tr>
</tbody>
</table>

**Contribution of VSLS to stratospheric bromine budget based on bottom-up approach**
CHBr$_3$ in the TTL (1993–2013)
Roles of oceanic emissions and atmospheric transport

2. Pacific
3. Indian Ocean
1. Central America

Analyze maxima, seasonality and long-term changes
CHBr$_3$ over Central America

17 km

- FLEXPART shows observed latitudinal and seasonal variations.
- FLEXPART slightly underestimates observed maxima in NH summer.
CHBr$_3$ over Central America – Emissions and Transport

17 km

Strong sources and fast vertical transport cause maxima over Central America.

Source distribution

Sea surface-to-TTL transit time

Source regions for Central America [%]

Transit time [days] at 17 km for JJA
Seasonality in TTL with maxima in NH summer is driven by:

- **Seasonality in surface-to-TTL transit time**
- **Seasonality in oceanic emissions**
CHBr$_3$ over Central/East Pacific

FLEXPART and aircraft measurements between 16 and 18 km

FLEXPART vs. ATTREX, Feb 2013

- FLEXPART shows observed latitudinal variations.
- Tropical maximum in central Pacific is too far south in simulations.
CHBr$_3$ over the Pacific

Source distribution mostly south of Equator

Transit time distribution roughly symmetric around Equator.
CHBr$_3$ over the Pacific

Source distribution mostly south of Equator

- FLEXPART simulations project Pacific maximum south of Equator as a result of strong sources here.
- More ship campaigns needed to confirm or improve source distribution in the tropical Pacific.
CHBr$_3$ over Indian Ocean – global maximum

Maximum CHBr$_3$ in NH summer over Bay of Bengal and Indian ocean.

But air masses are youngest over the West Pacific (white lines show sea surface-to-TTL transit time distribution).
CHBr₃ over Indian Ocean – Sources

Maximum is caused by strong sources in Arabian sea and Bay of Bengal during NH summer.

→ see talk by Alina Fiehn Wednesday morning for detailed evaluations of VSLS emission and transport processes in the Asian monsoon region
Interannual and long-term changes

Overall increase in TTL CHBr$_3$ abundance due to increasing emissions and decreasing transit times. Strongest trend above the Indian Ocean.
CHBr$_3$ in the TTL – Summary

1. Coinciding sources and fast vertical transport
   - Maximum over Central America
   - Pronounced seasonal cycle.

3. Strong sources in Arabian sea and Bay of Bengal
   - Global Maximum
   - Pronounced seasonal cycle
   - Strongest long-term changes

2. Shortest sea surface-to-TTL transit times over Pacific
   - Maximum south of Equator as a result of the source distribution (based on only a few measurements)
   - Weak seasonal cycle but pronounced interannual variability