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

S. Tegtmeier[1]

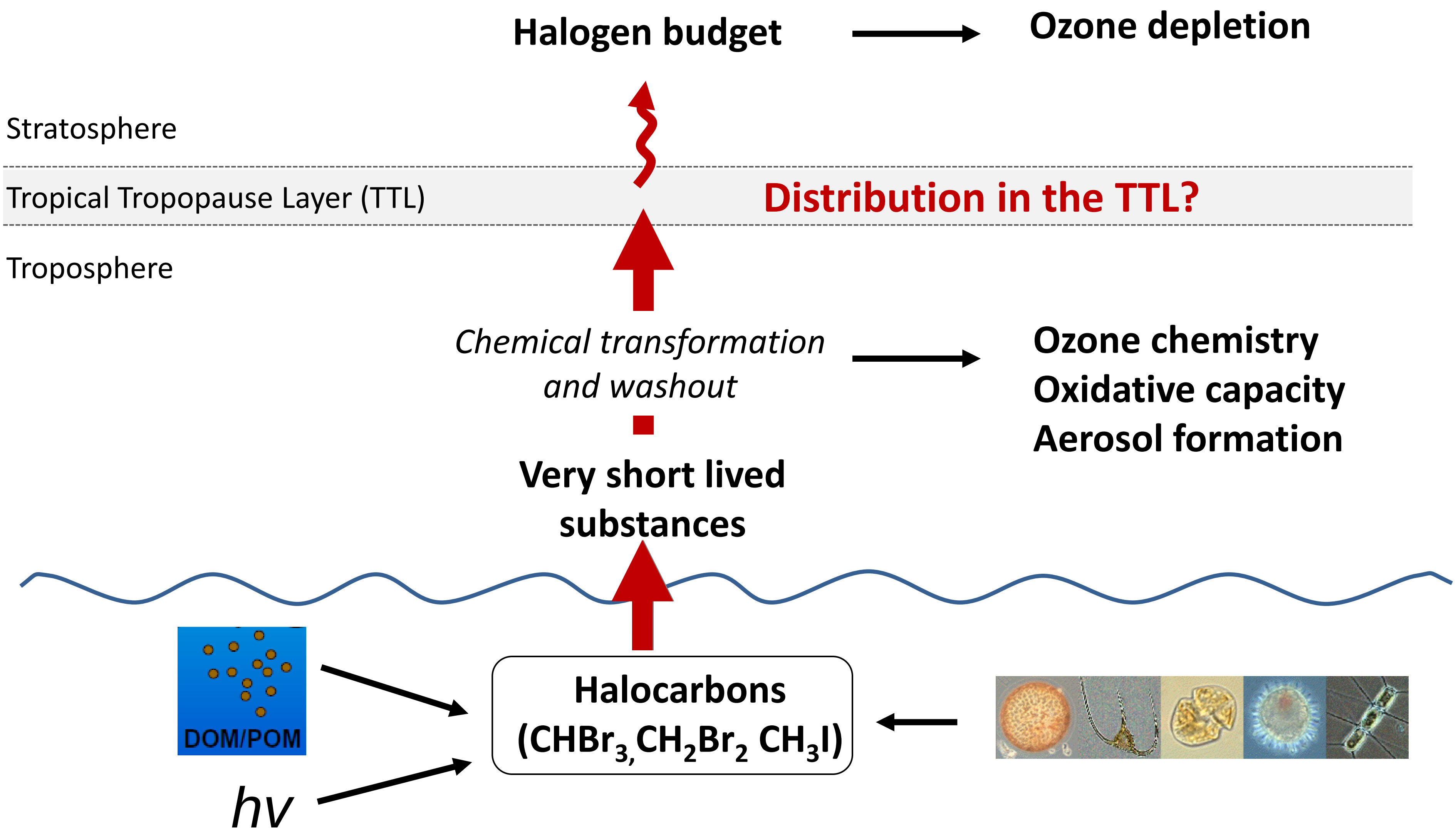
K. Krüger[2], B. Quack[1], E. Atlas[3], F. Ziska[1]

[1]GEOMAR Helmholtz Centre for Ocean Research Kiel, Kiel, Germany
[2]Department of Geosciences, University of Oslo, Oslo, Norway
[3]Rosenstiel School of Marine and Atmospheric Science, University of Miami, Florida, USA

GEFÖRDERT VOM
Bundesministerium für Bildung und Forschung

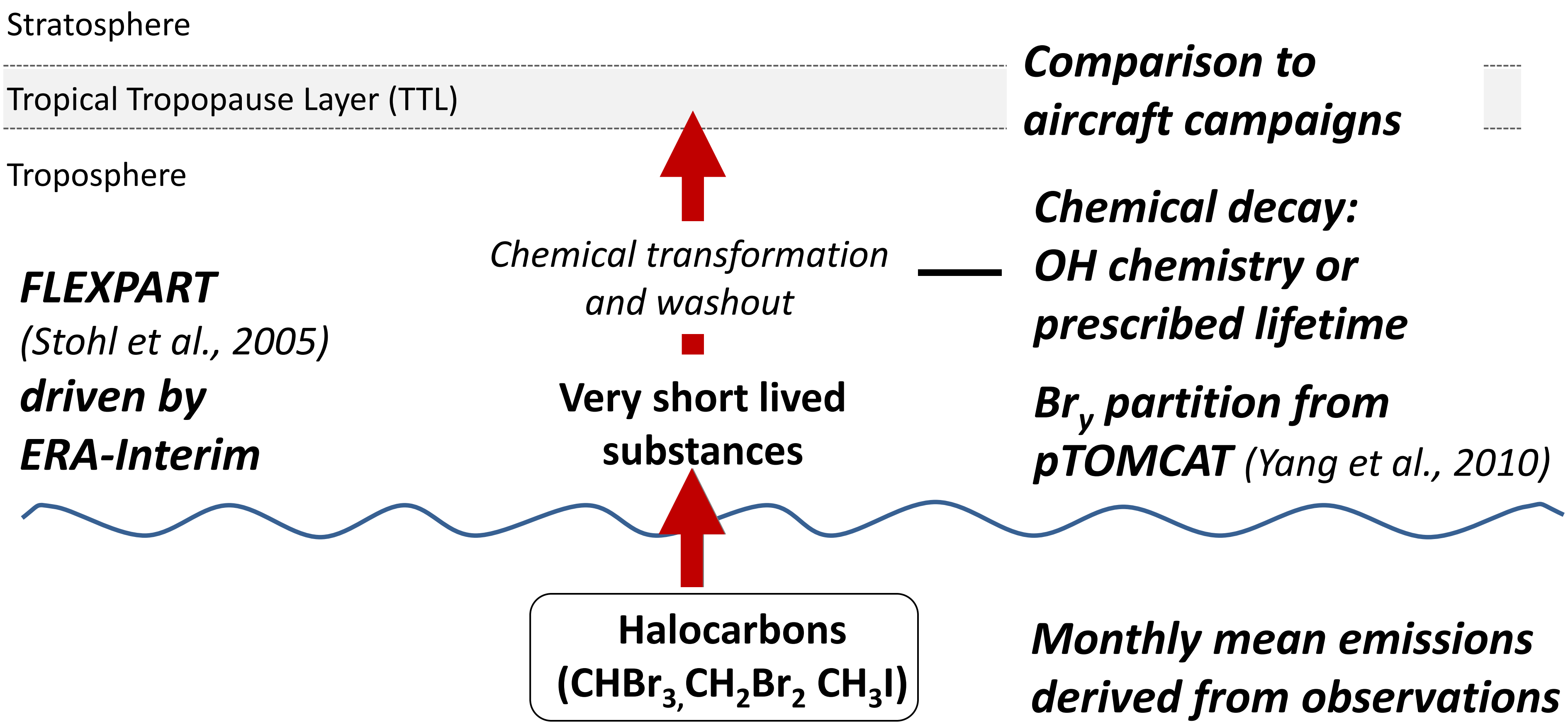
GEOMAR

Motivation

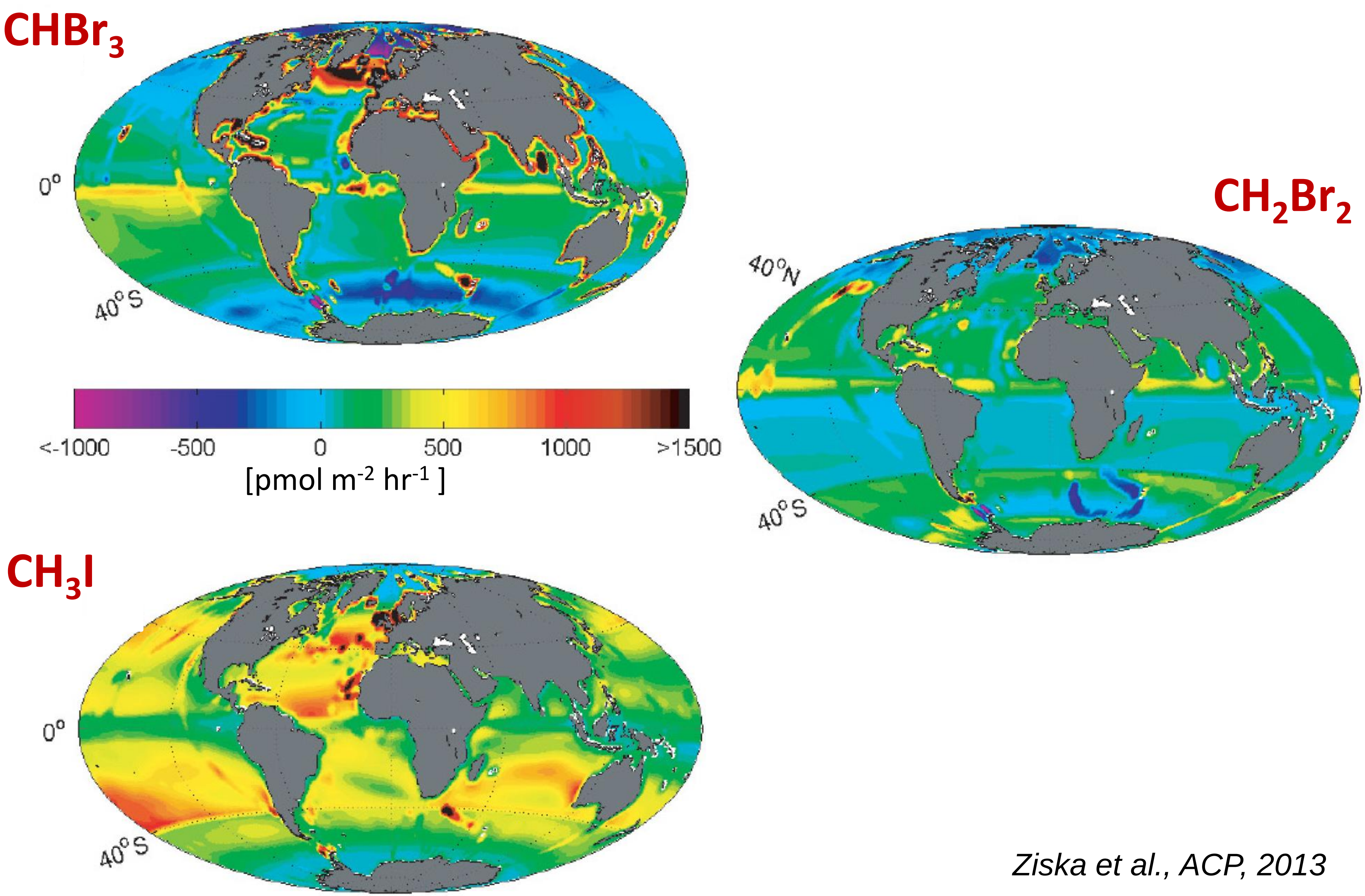

Halogen budget
Ozone depletion
Stratosphere
Tropical Tropopause Layer (TTL)
Distribution in the TTL?
Troposphere
Chemical transformation and washout
Ozone chemistry
Oxidative capacity
Aerosol formation
Very short lived substances
DOM/POM
Halocarbons
($CHBr_3$, CH_2Br_2 CH_3I)
hv

Method

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



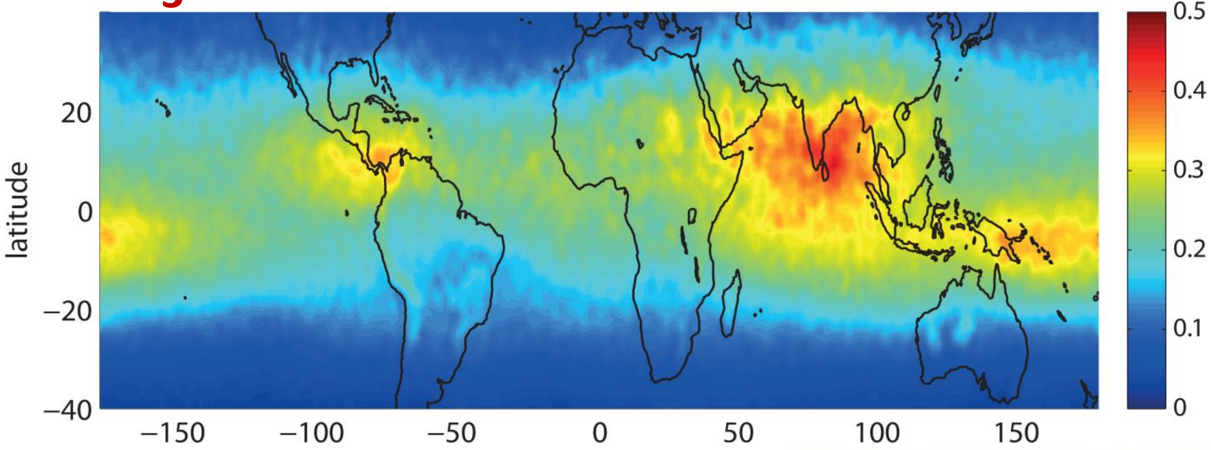
Input for FLEXPART: global VSLS emissions (Ziska climatology)



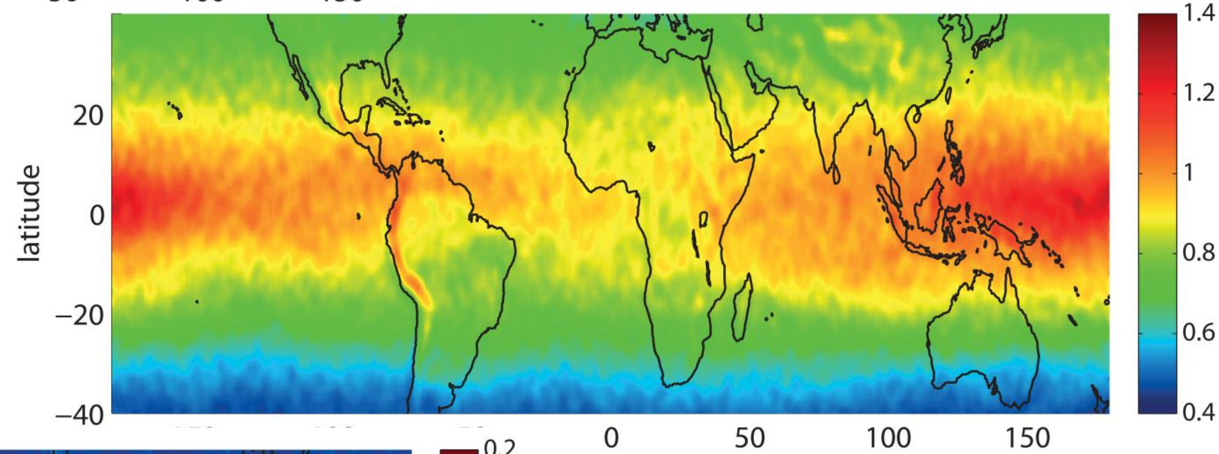
Ziska et al., ACP, 2013

Results: Annual mean VMR @ 17 km [ppt] 2008-2010

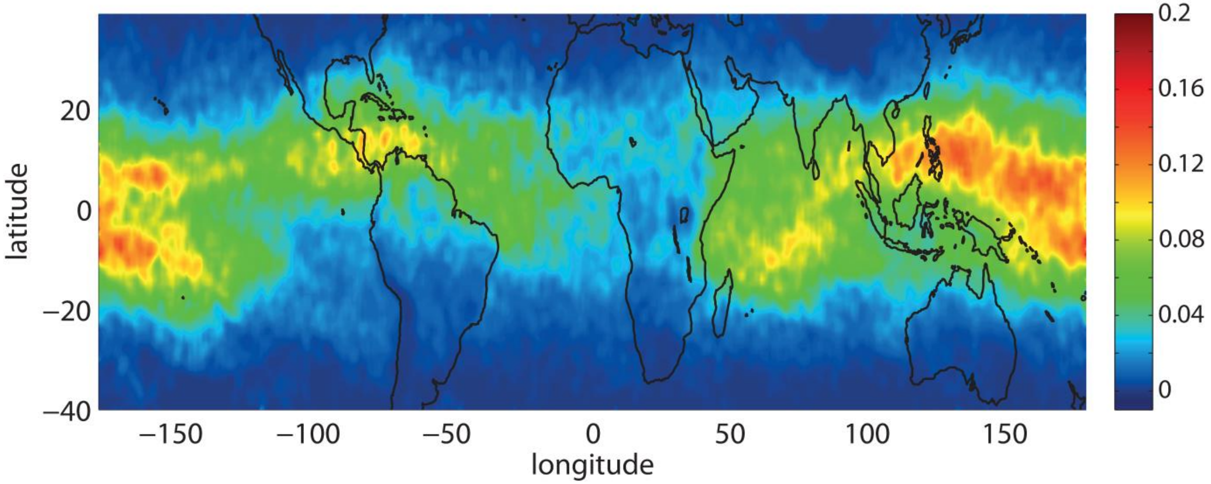
$CHBr_3$



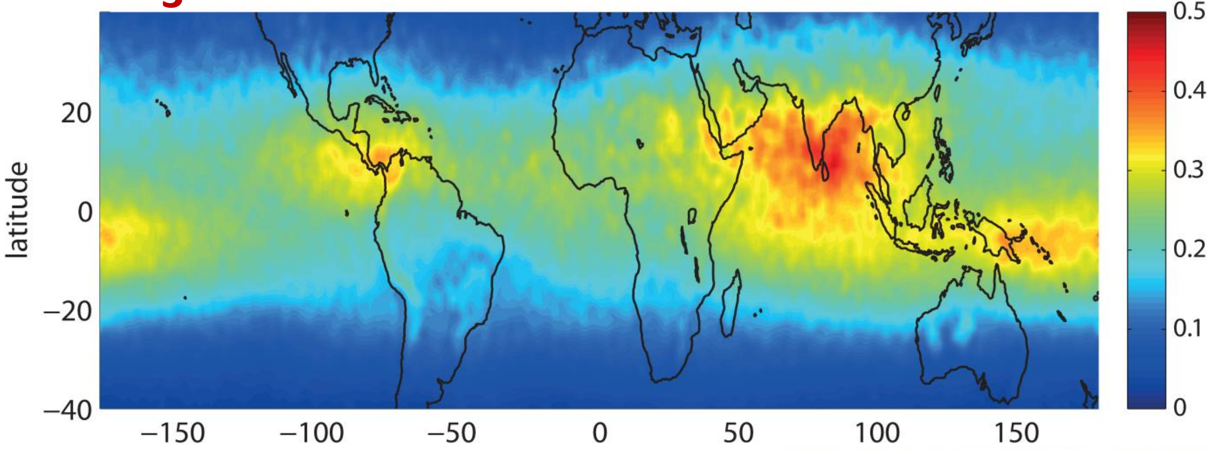
CH_2Br_2



CH_3I

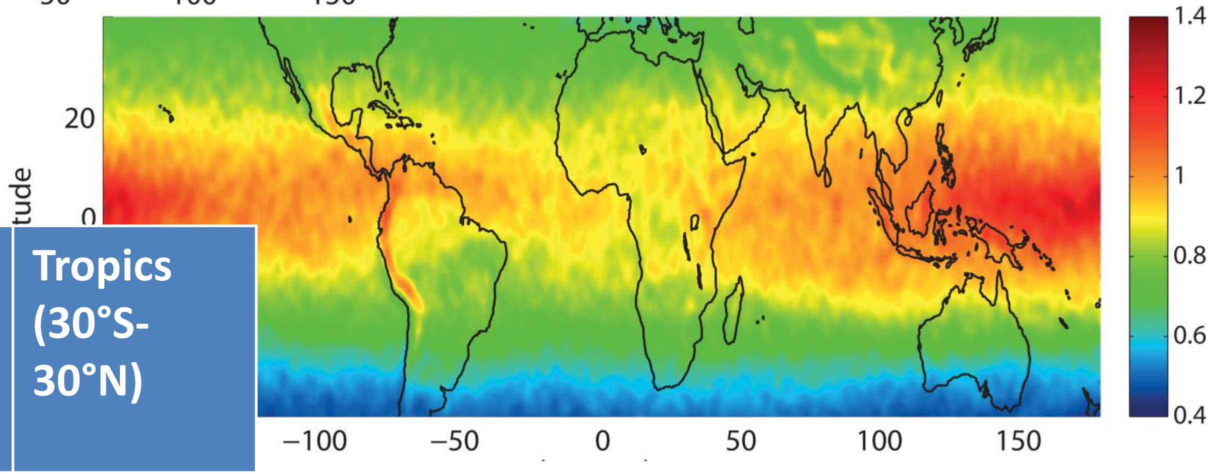


$CHBr_3$



Results:
Annual mean VMR @ 17 km [ppt] 2008-2010

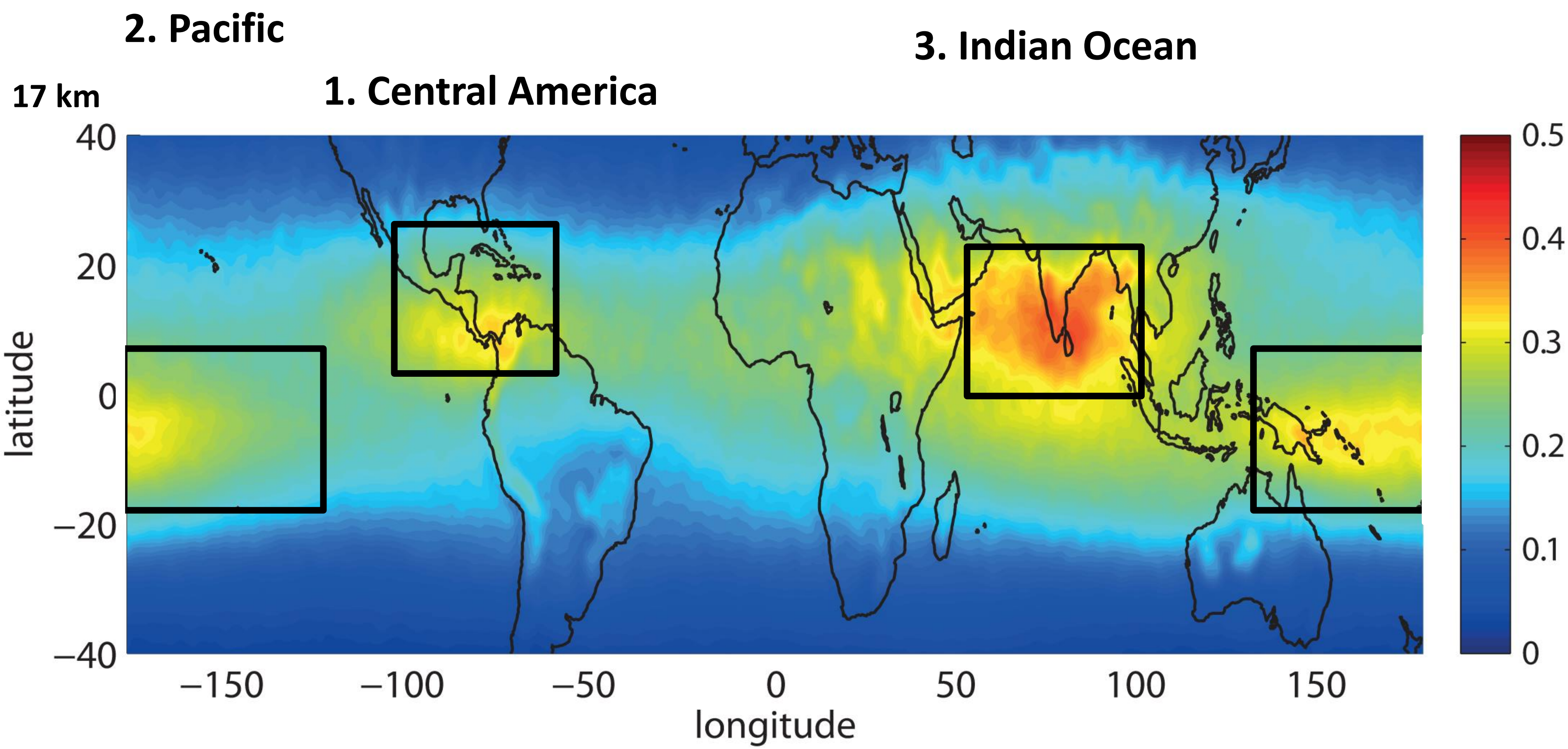
CH_2Br_2



Br [ppt] Based on …	Inner tropics (10°S-10°N)	Tropics (30°S-30°N)
$CHBr_3$ SG	0.8	0.6
$CHBr_3$ SG+PG	1.3	1.1
CH_2Br_2 SG	2.0	1.7
CH_2Br_2 SG + PG	2.1	2.0
All	**3.4**	**3.1**

Contribution of VSLS to stratospheric bromine budget based on bottom-up approach

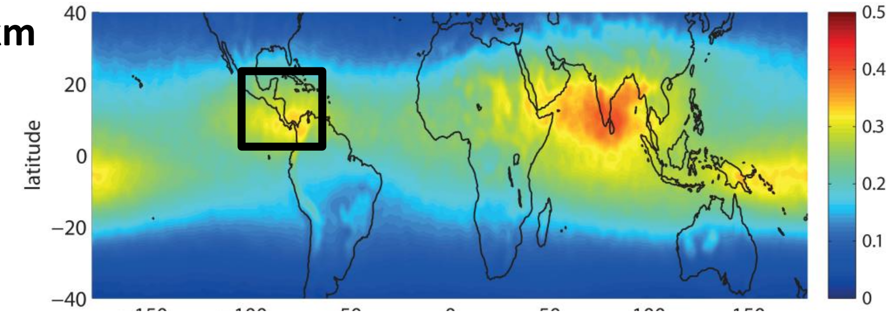
$CHBr_3$ in the TTL (1993–2013)
Roles of oceanic emissions and atmospheric transport



Analyze maxima, seasonality and long-term changes

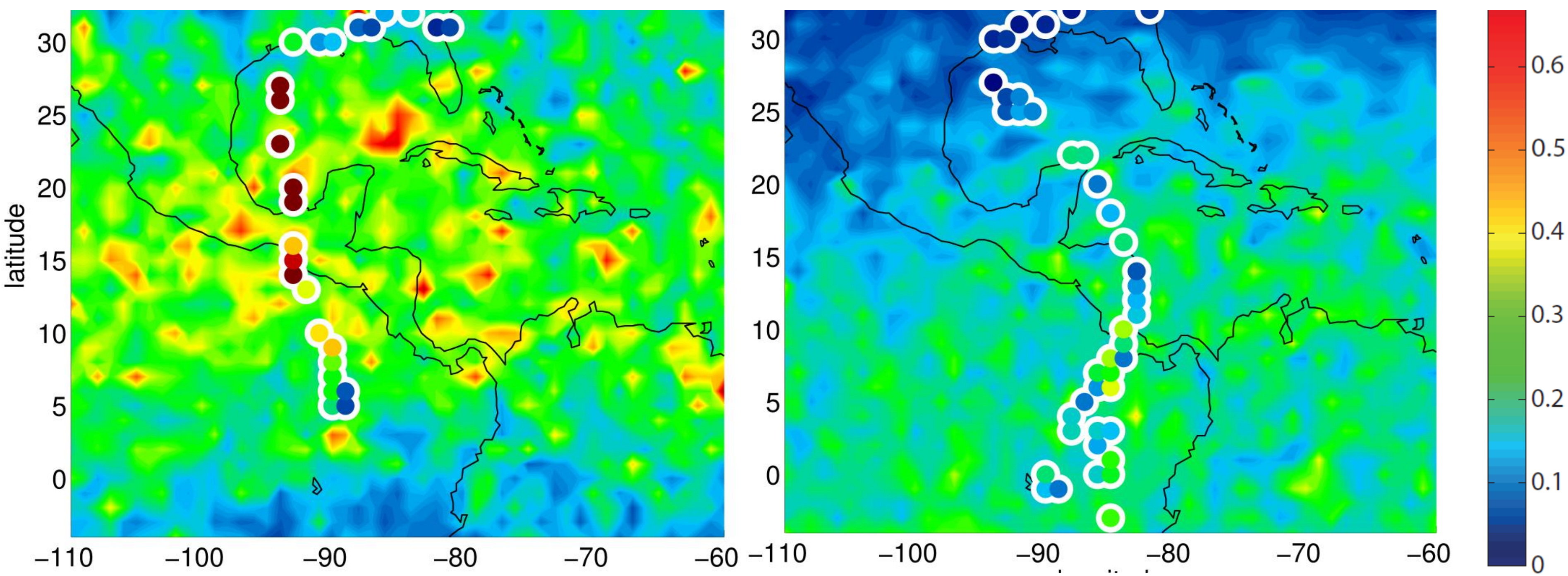
$CHBr_3$ over Central America

17 km



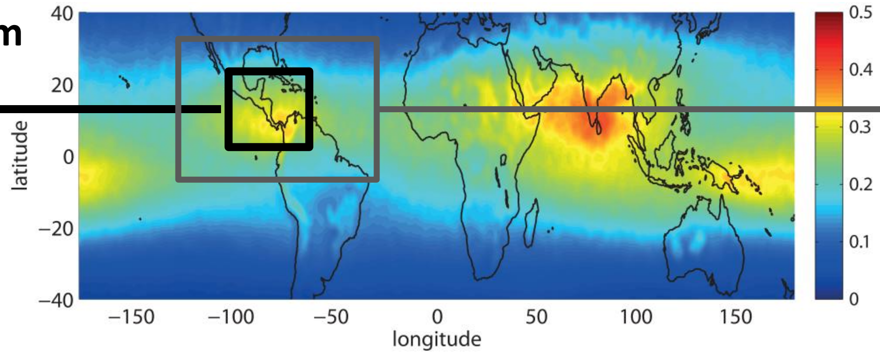
FLEXPART and
aircraft measurements
between 16 and 18 km

- **FLEXPART shows observed latitudinal and seasonal variations.**
- **FLEXPART slightly underestimates observed maxima in NH summer.**



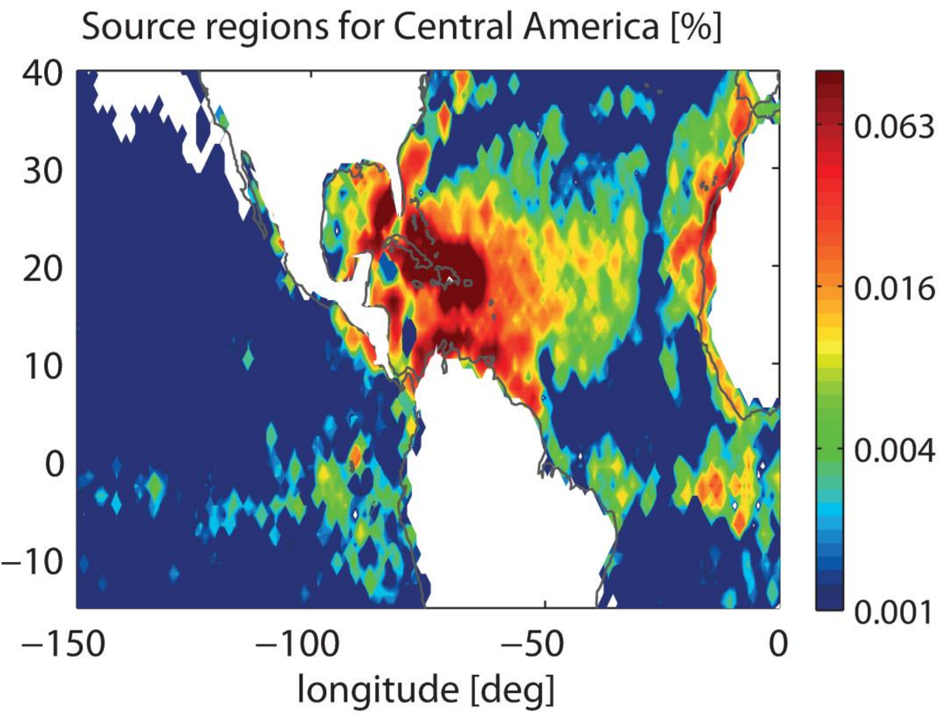
$CHBr_3$ over Central America – Emissions and Transport

17 km

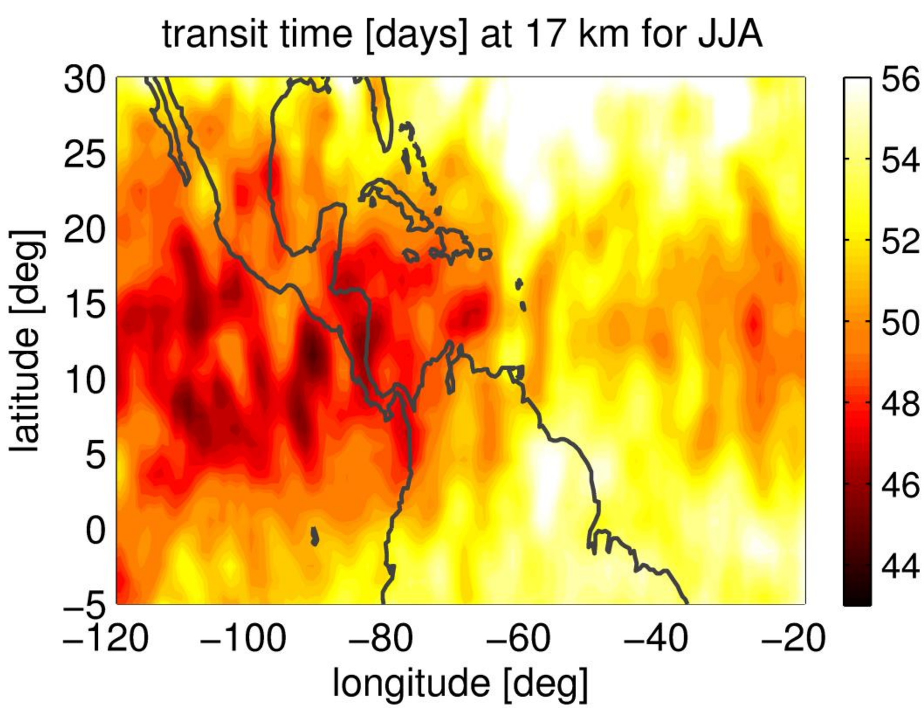


Strong sources and fast vertical transport cause maxima over Central America.

Source distribution

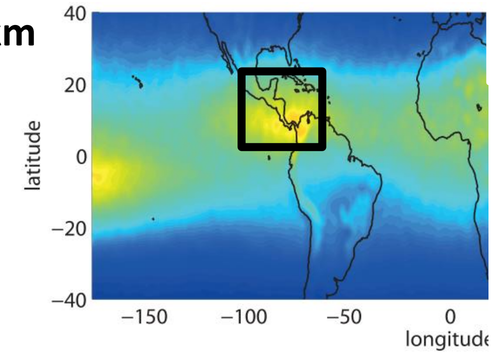


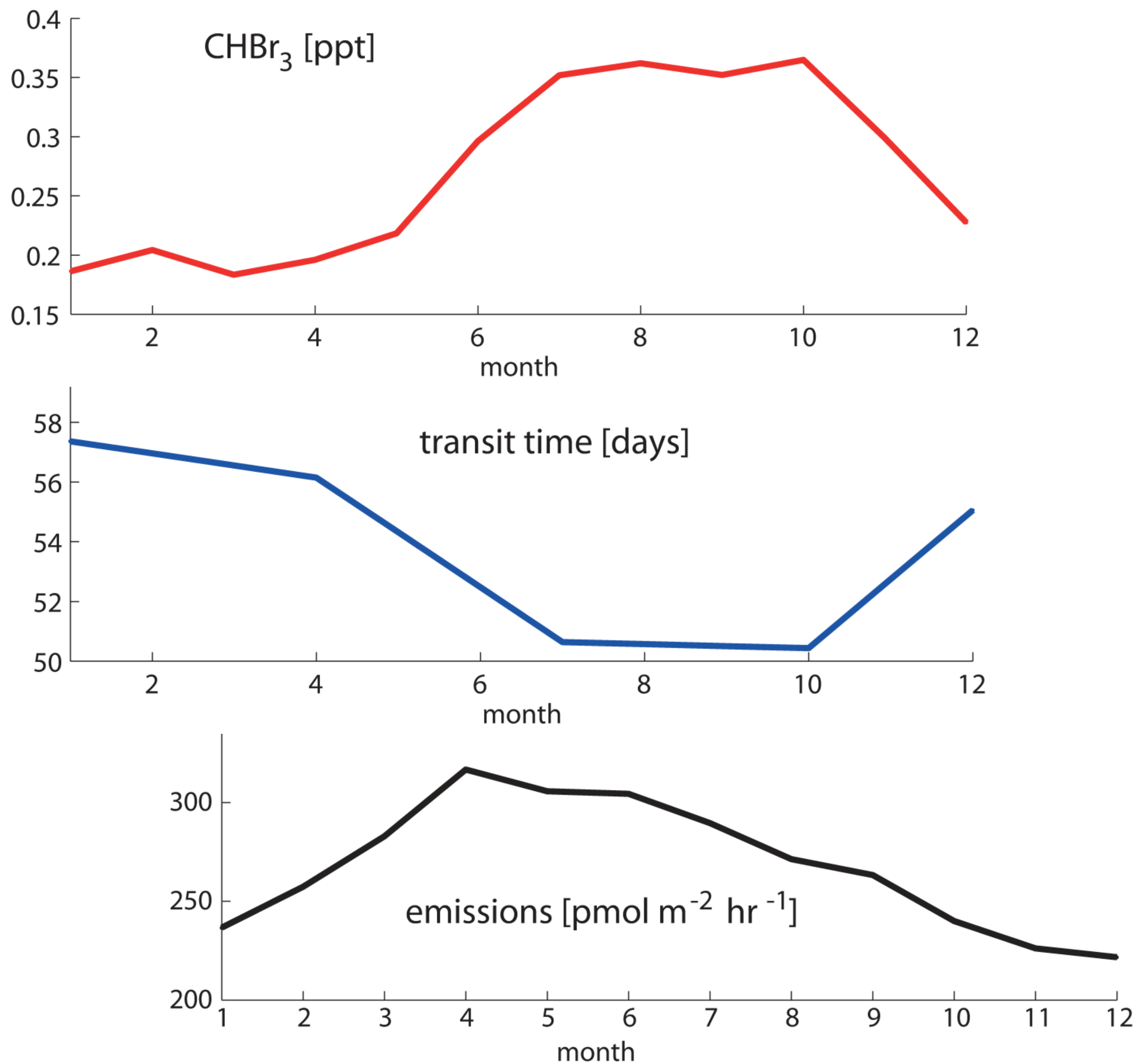
Sea surface-to-TTL transit time



$CHBr_3$ over Central America – Seasonality

17 km

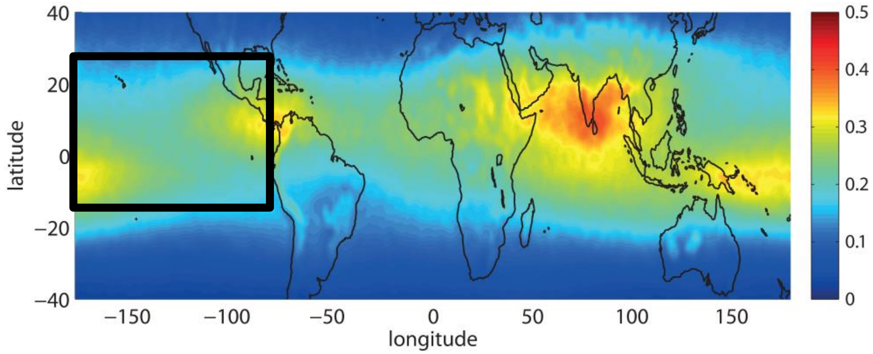




Seasonality in TTL with maxima in NH summer is driven by:

- **Seasonality in surface-to-TTL transit time**
- **Seasonality in oceanic emissions**

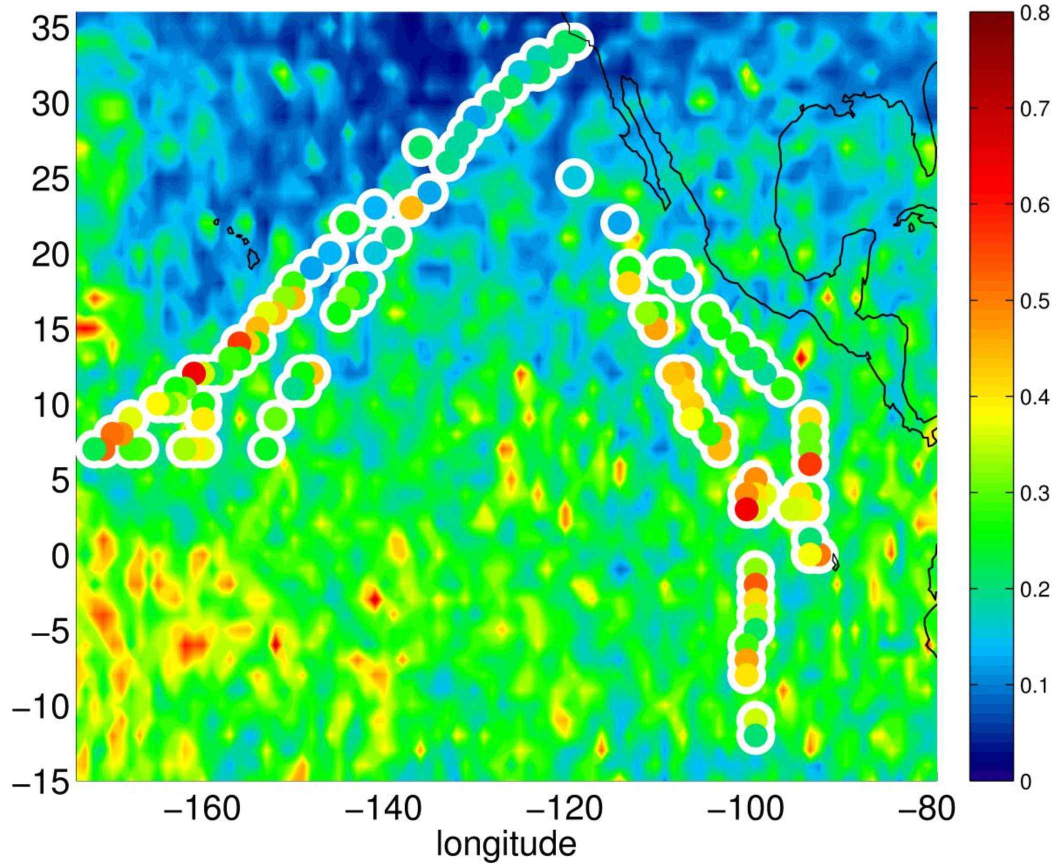
$CHBr_3$ over Central/East Pacific



FLEXPART and aircraft measurements

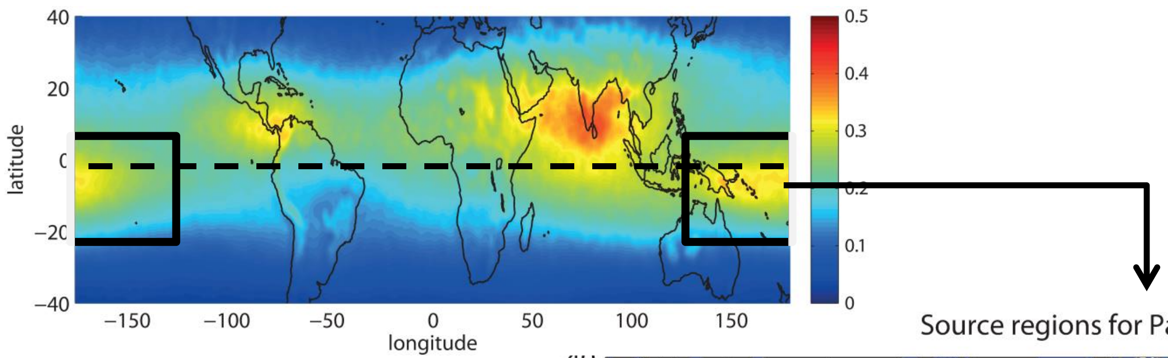
between 16 and 18 km

FLEXPART vs. ATTREX, Feb 2013

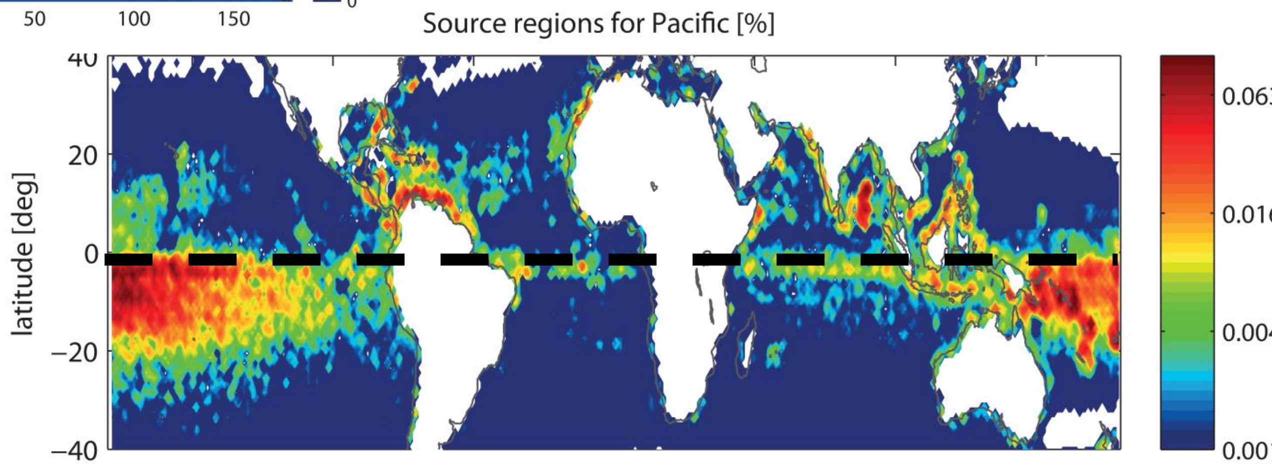


- **FLEXPART shows observed latitudinal variations.**
- **Tropical maximum in central Pacific is too far south in simulations.**

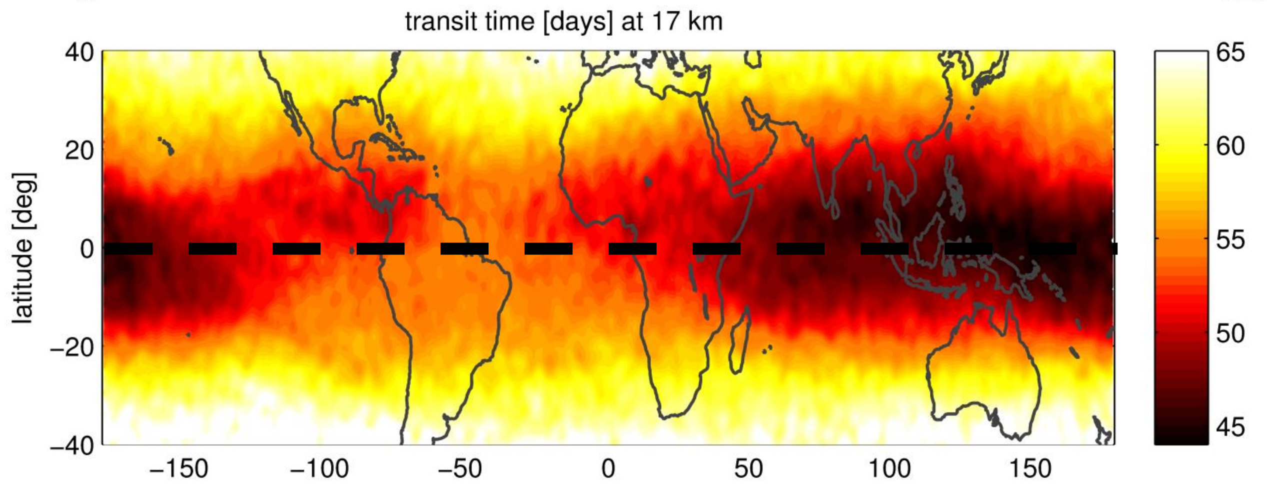
$CHBr_3$ over the Pacific



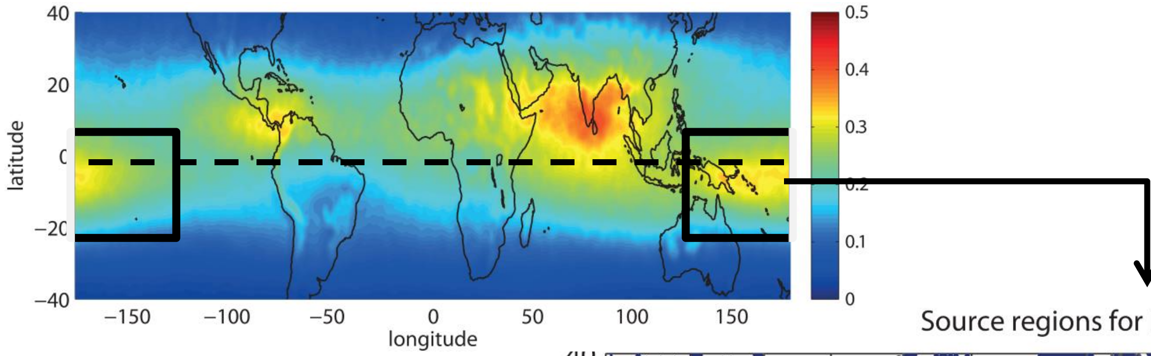
Source distribution mostly south of Equator



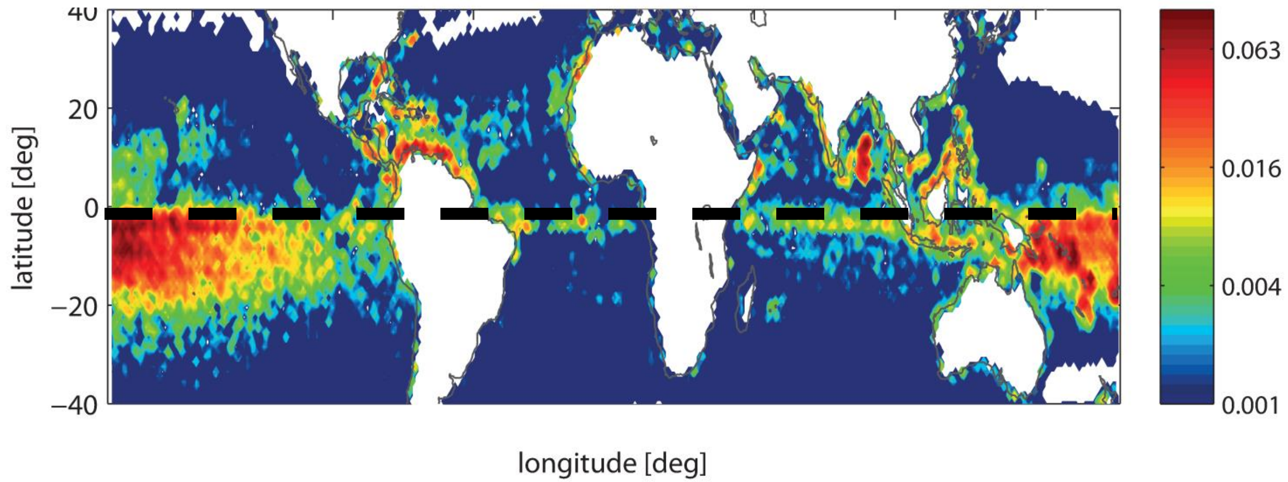
Transit time distribution roughly symmetric around Equator.



$CHBr_3$ over the Pacific



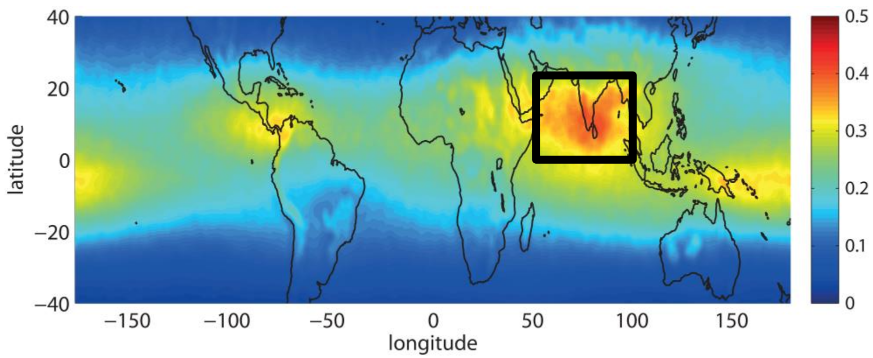
Source regions for Pacific [%]



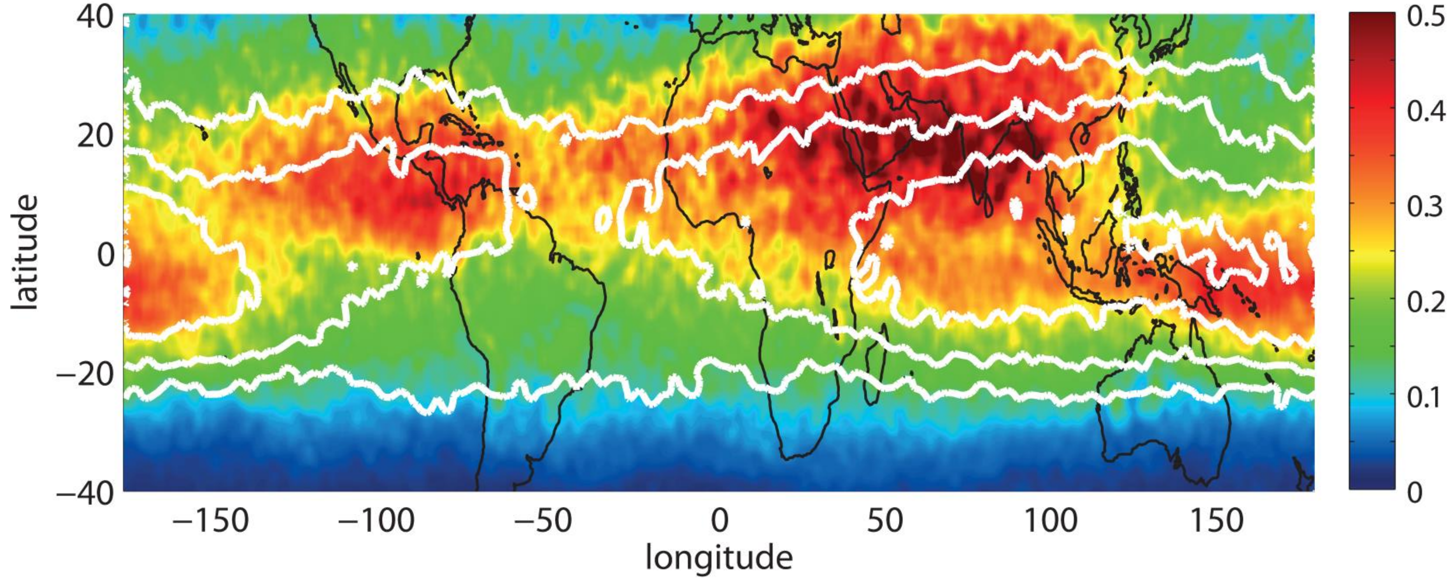
Source distribution mostly south of Equator

- **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



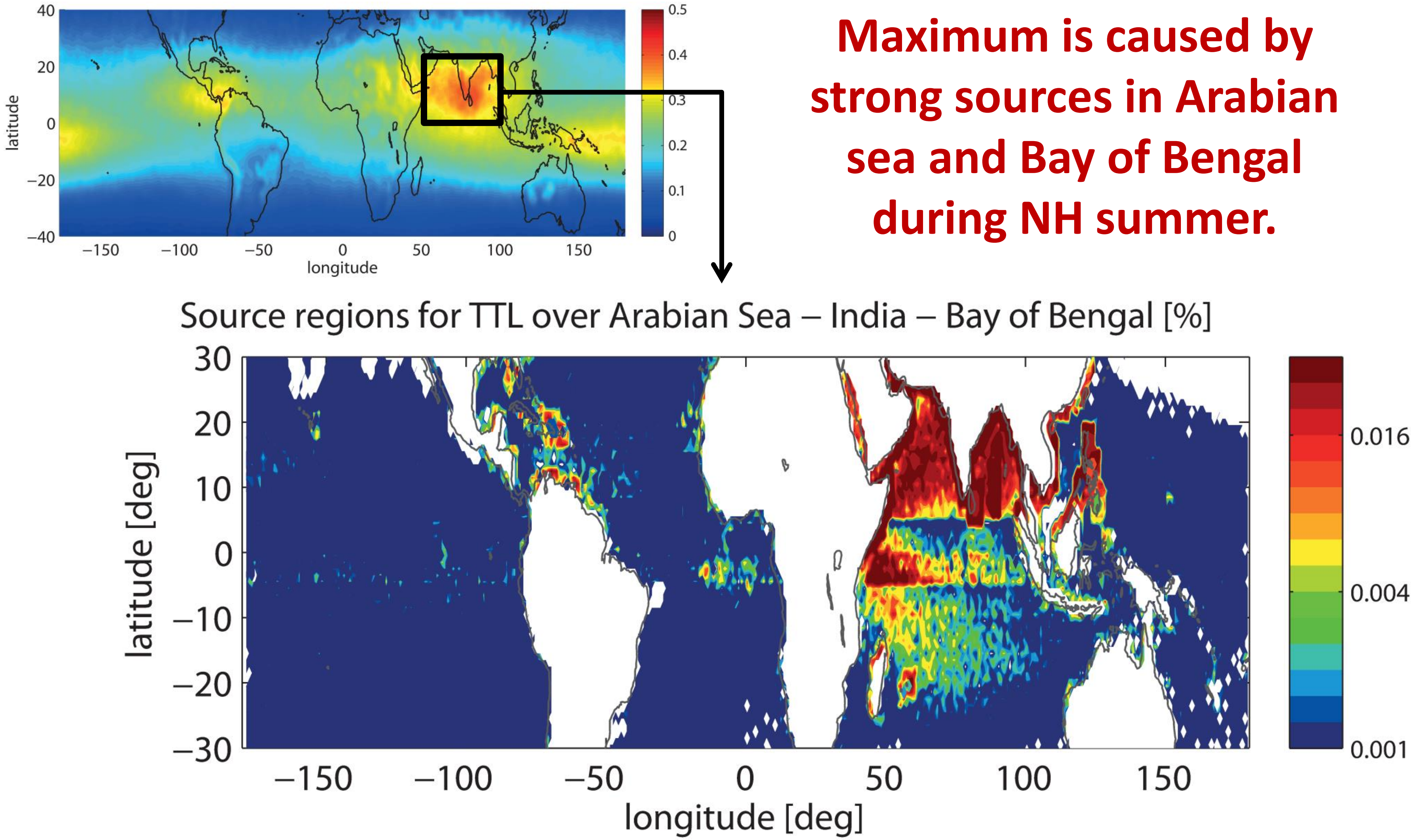
Maximum $CHBr_3$ in NH summer over Bay of Bengal and Indian ocean.



But air masses are youngest over the West Pacific

(white lines show sea surface-to-TTL transit time distribution).

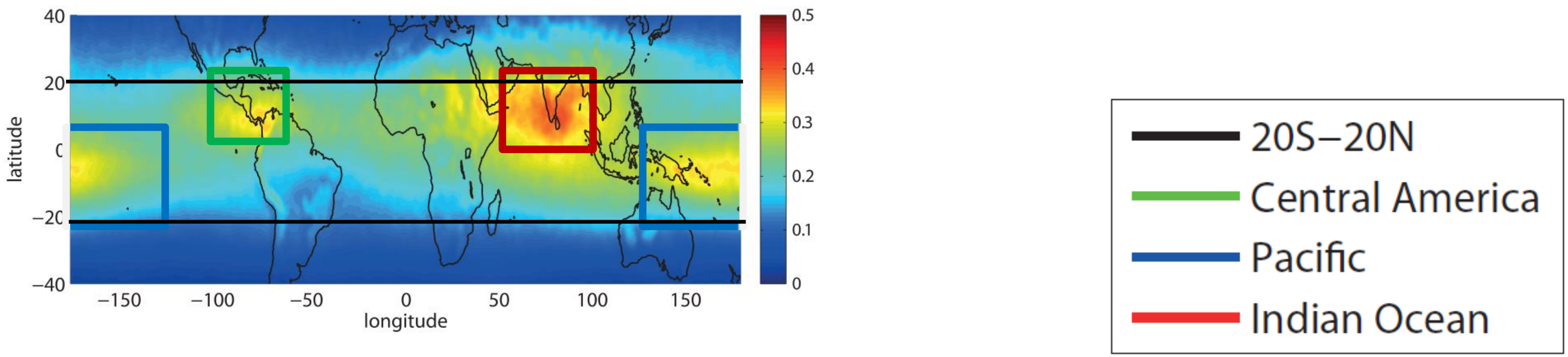
$CHBr_3$ over Indian Ocean – Sources



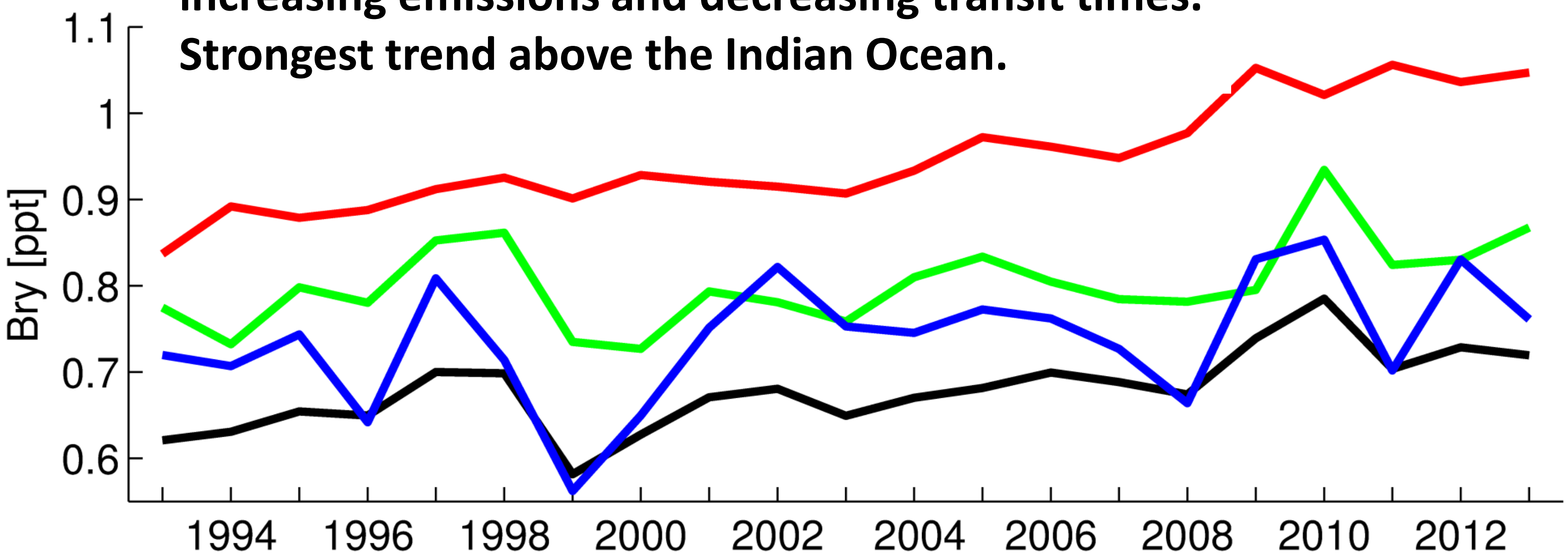
Maximum is caused by strong sources in Arabian sea and Bay of Bengal during NH summer.

→ 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.



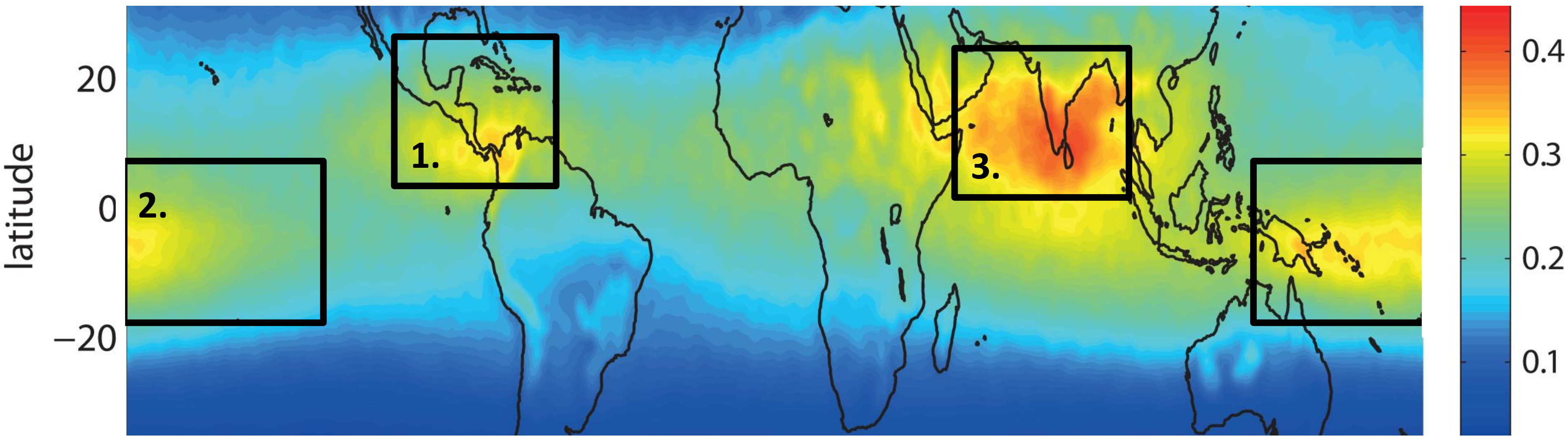
$CHBr_3$ in the TTL – Summary

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