Convective transport of very-short-lived bromocarbons to the stratosphere

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Introduction

- Recent estimates of very short lived bromocarbons (VSLB) to stratospheric bromine (Br_y^{VSLB})
  - Balloon and satellite based: 6 (3-8.4) ppt [Montzka and Reimann, 2011)
  - Model estimates: 4.8-7 ppt

- Remaining uncertainties:
  - Satellite estimates: varying vertical sensitivities and sampling altitudes
  - Balloon estimates: the j(BrONO2) / (k(BrO+NO2) ratio used
  - Model estimates: various simplification of emission, transport, wet scavenging processes

- Is Br_y^{VSLB} sensitive to convection intensity?
  - Sinnhuber & Folkins (2006): Different washout rates -> 0.5-3 ppt range in Br_y^{CHBr3}.
  - Aschmann et al. (2009): Different washout rates -> 1.6-3 ppt range in Br_y^{CHBr3}.
  - Liang et al. (2010): Convective scavenging leads to ~0.2 ppt (4%) difference in Br_y^{VSLB}.
  - Aschmann et al. (2011): Intensified convection leads to higher source gas injection, but more washout of product gases, which cancel off with each other.

Model description

- Model simulations are run with the NASA Goddard GEOS Chemistry Climate Model (GEOSCCM) V2 – GEOS-5 coupled with standard stratospheric chemistry scheme and interactive $CHBr_3$ and CH_2Br_2 chemistry.

- The $CHBr_3$ and CH_2Br_2 chemistry scheme:
  - Spatially resolved $CHBr_3$ and CH_2Br_2 emissions at the surface, described in Liang et al. (2010).
  - Photochemical removal by photolysis and OH (fully interactive stratospheric OH + tropospheric climatology).
  - Speciation for sources and inorganic products, HBr, HOBr, $BrONO_2$, BrCl, Br, BrO, Br_2
  - Detailed washout and rainout scheme for HBr, HOBr and $BrONO_2$ in large-scale and convective precipitation, with consideration of re-evaporation.
  - No heterogeneous chemistry, but impact should be small (Aschmann and Sinnhuber, 2013).

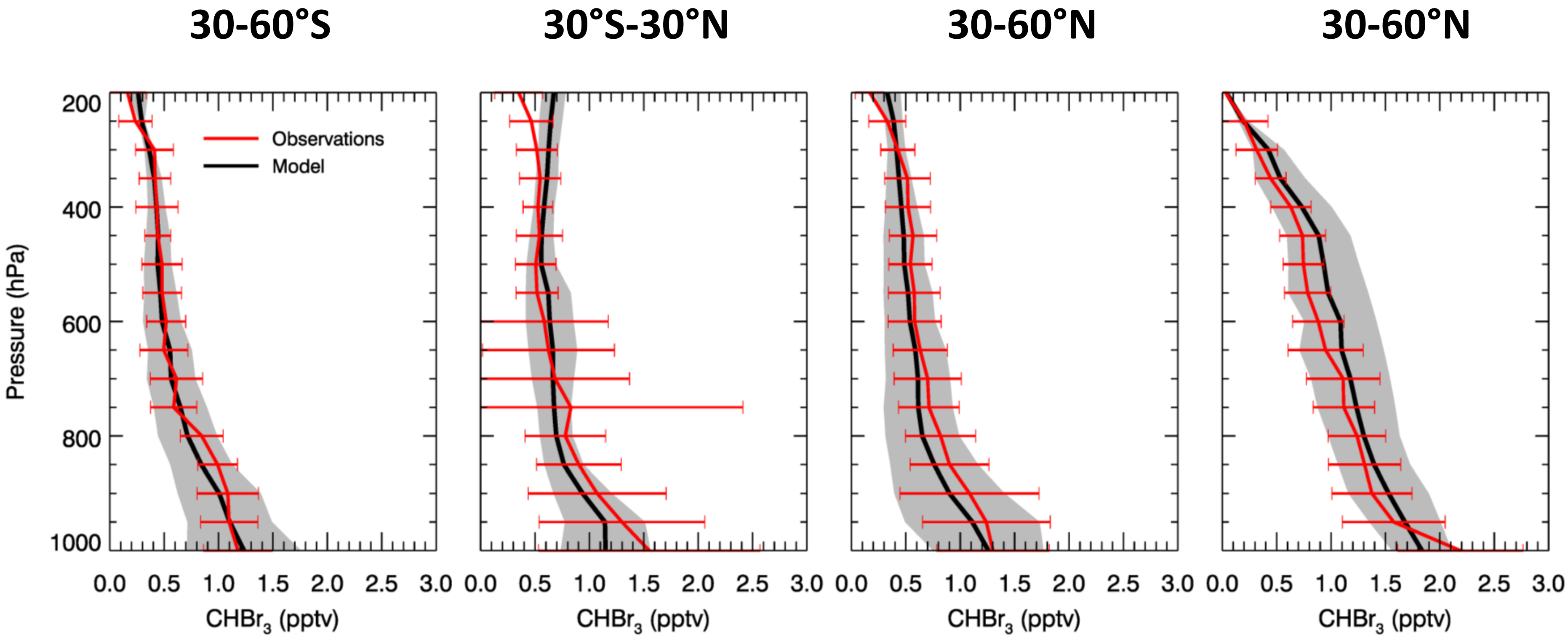
Simulation description

- Two 50-year (1960-2010) simulations to quantify the contribution of VSLB to Br_y:
  - A baseline simulation (without VSLB) – $\boldsymbol{R_{BASE}}$
  - A parallel simulation with VSLB – $\boldsymbol{R_{VSLB}}$

- Two 30-year (1980-2010) sensitivity simulations to examine the impact of convection strength on Br_y^{VSLB}:
  - A run with Minimum convection condition – $\boldsymbol{R_{MINCNV}}$
  - A run with Maximum convection condition -- $\boldsymbol{R_{MAXCNV}}$

By varying five convective parameters in RAS that impact the strength of deep convection, clouds, convective condensate and re-evaporation [Ott et al., 2011].

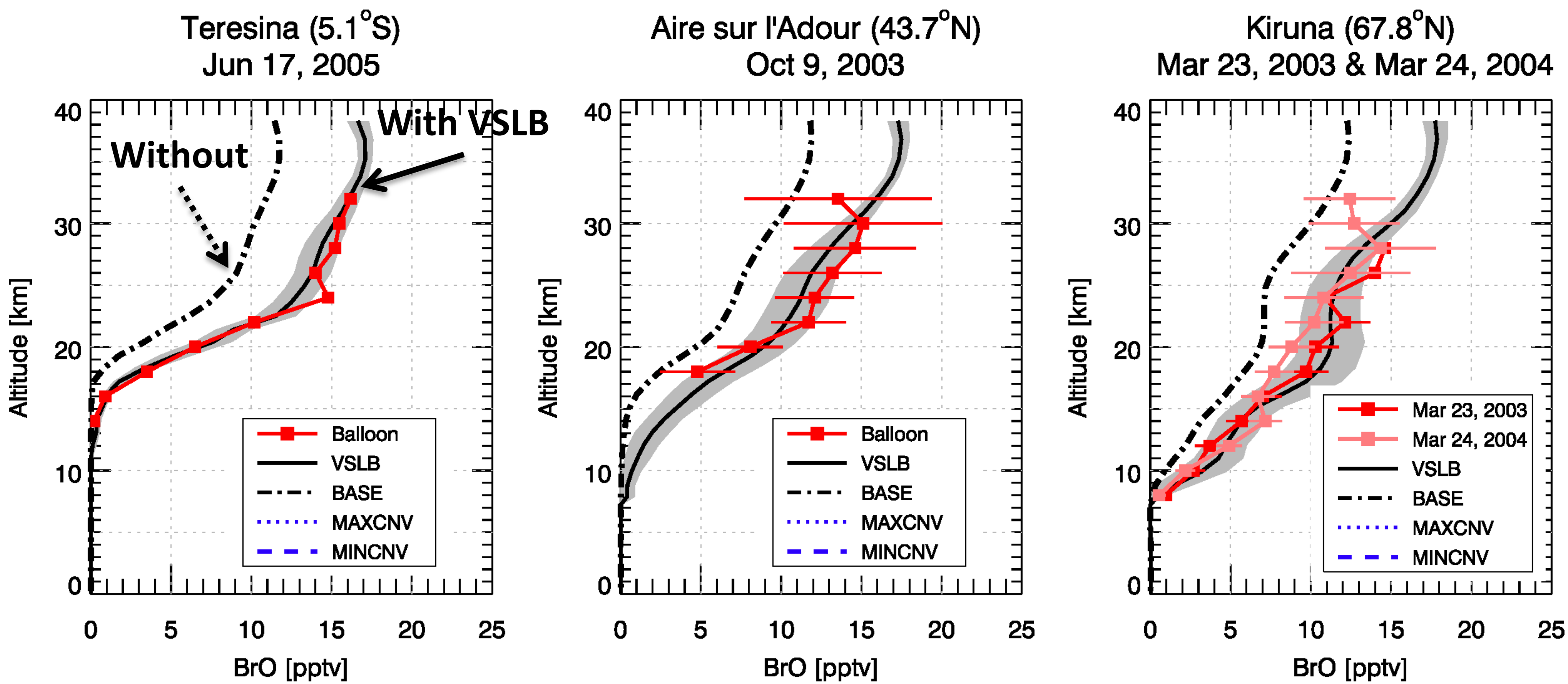
Modeled $CHBr_3$ vs. Observations



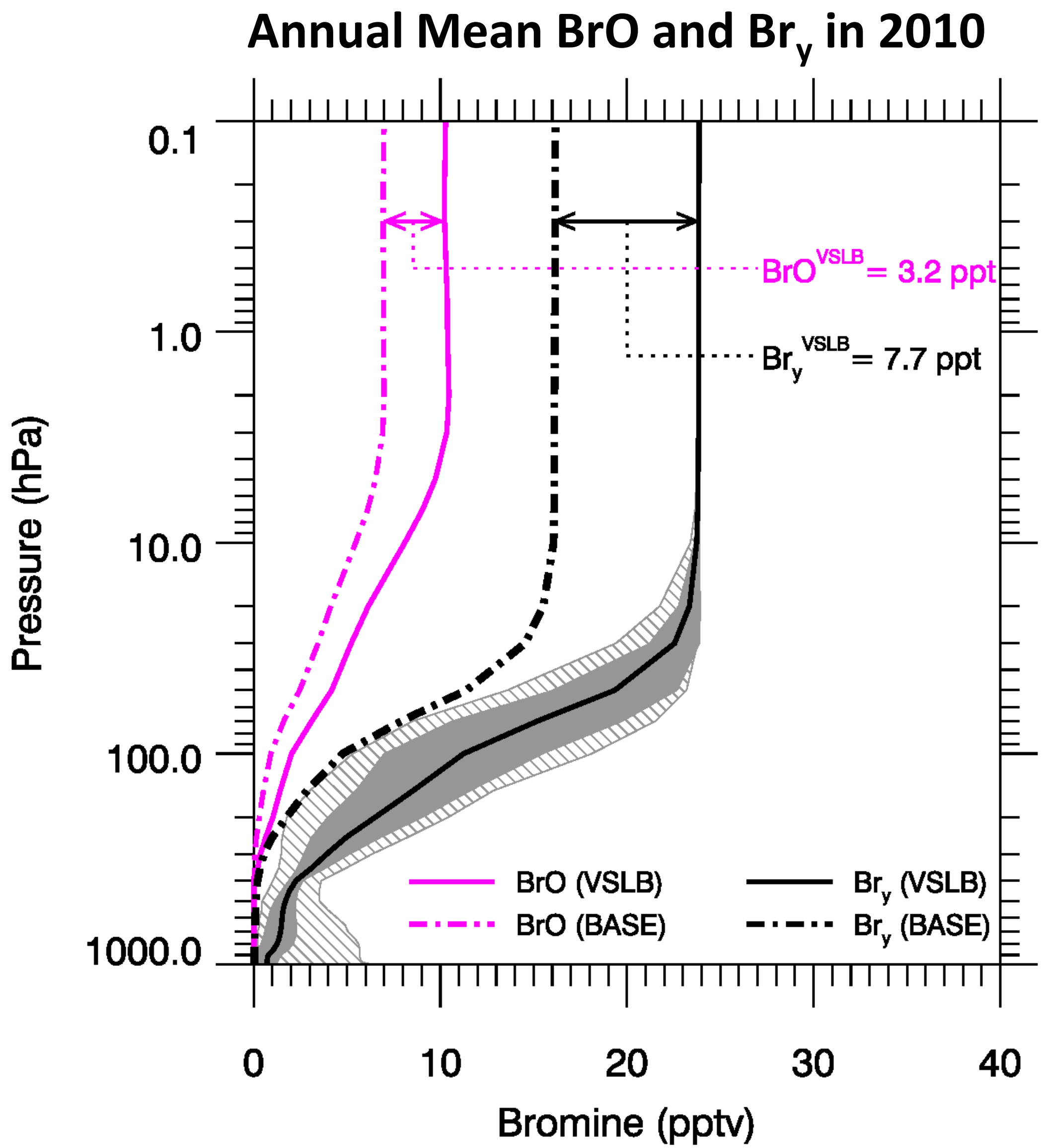
Observed profiles are compiled using WAS observations from eight recent NASA field campaigns.

Modeled BrO vs. DOAS Balloon Observations

Tropics | **Mid Latitudes** | **High Latitudes**



VSLB contribution to BrO and Br_y



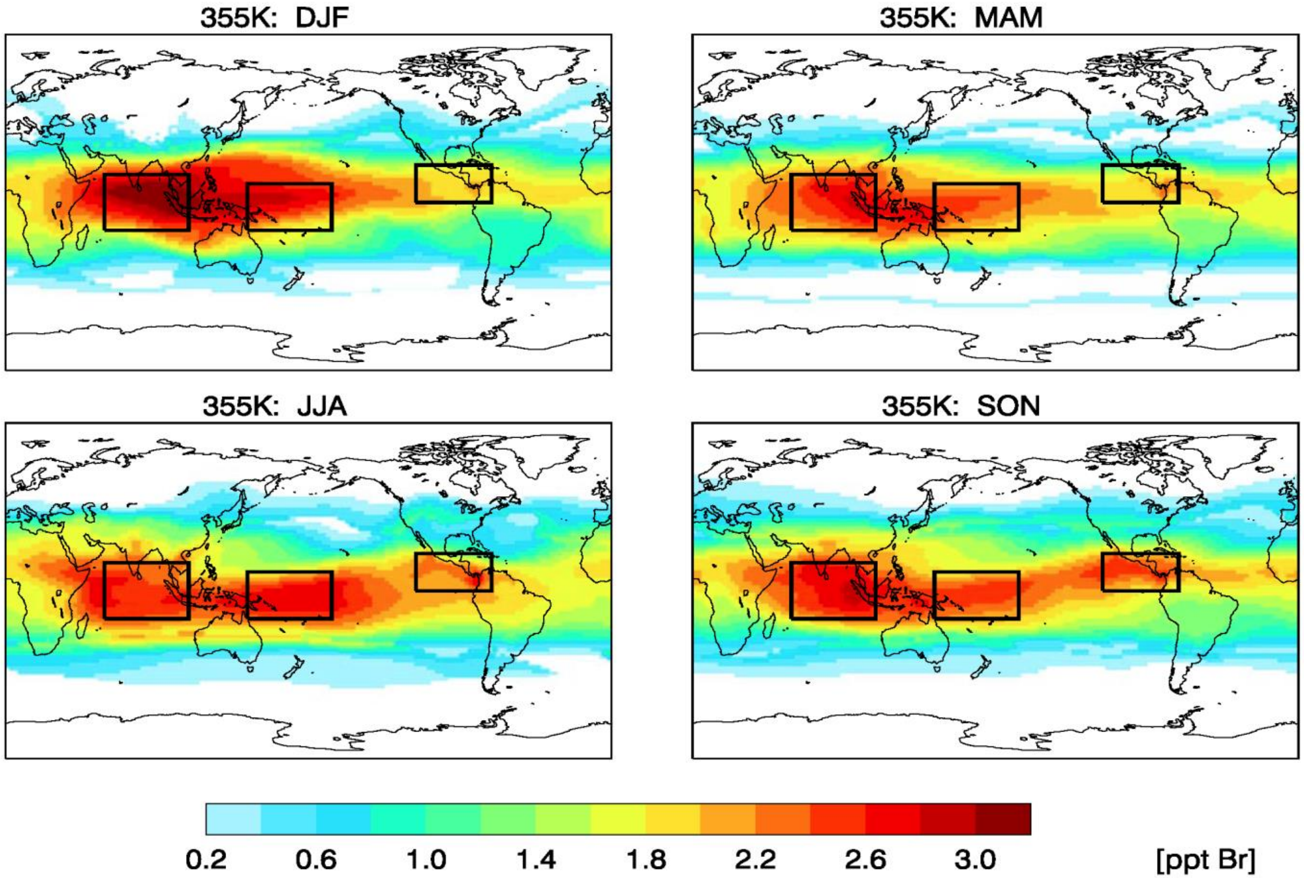
Inclusion of VSLB in the interactive scheme adds an average 7.7 ppt to stratospheric Br_y, ~50% higher than the ~5 ppt in Liang et al. (2010) when all Br_y^{VSLB} is treated as an soluble idealized tracer.

➔ *Partition between soluble and insoluble inorganic forms is important for model estimate of* Br_y^{VSLB} .

Convective lofting of VSLS to the Stratosphere: When and Where?

Seasonal Mean $CHBr_3$ (x 3) just below tropopause (355K)

Note 365 K marks the zero radiative heating.



Three active convective lofting regions:

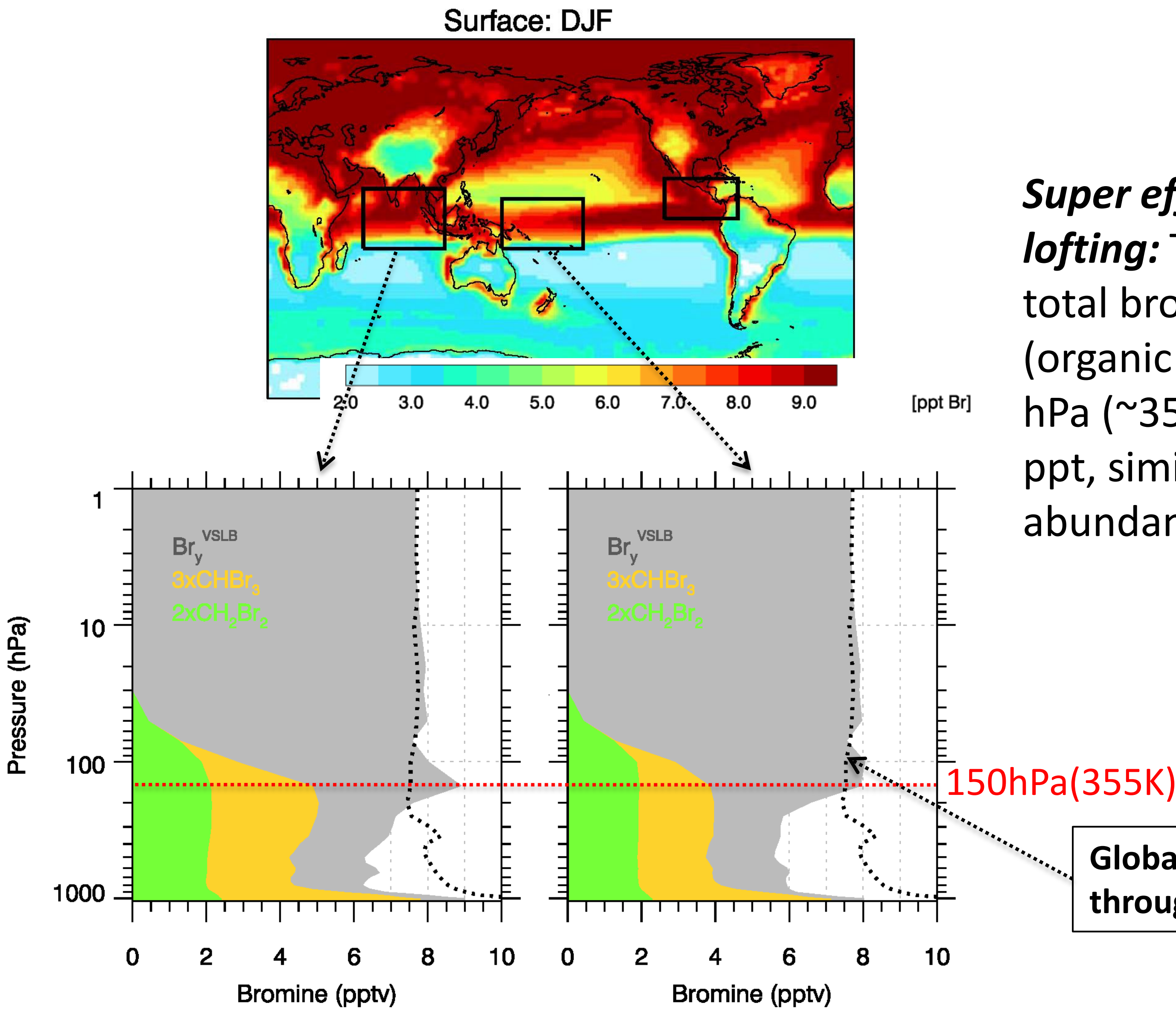
1) The tropical Indian ocean (boreal winter).

   (*Levine et al. 2007*)

2) **The western Pacific warm pool (all year long). – Targeting region for CONTRAST.**

3) The Pacific coast of Mexico (boreal fall).

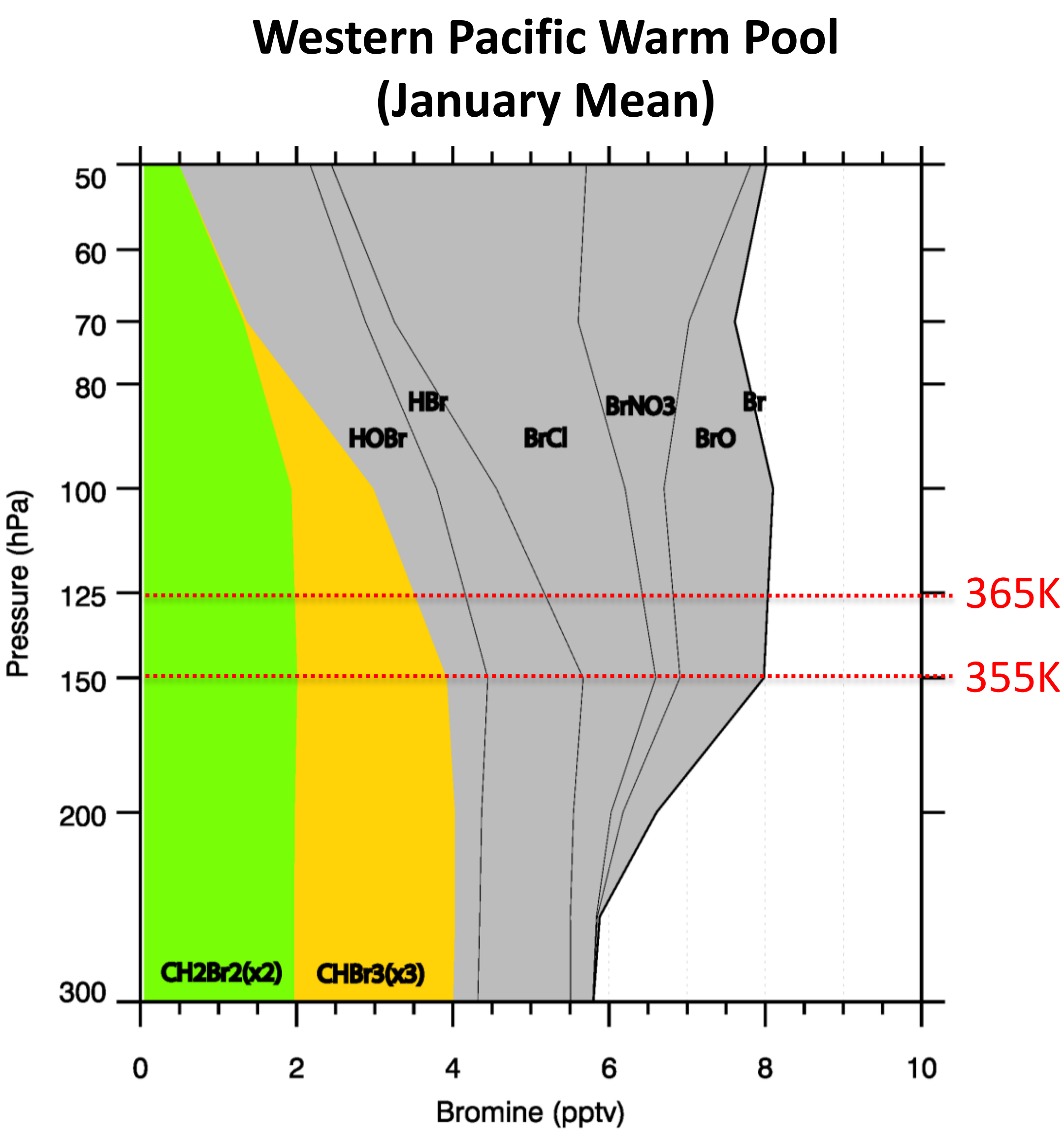
Surface to Stratosphere transport



Super efficient convective lofting: The mixing ratio of total bromine from VSLB (organic + inorganic) at 150 hPa (~355K) is about 8.0 - 8.5 ppt, similar to the surface abundance.

Global Mean Br_y^{VSLB} ~ 7.7 ppt throughout stratosphere.

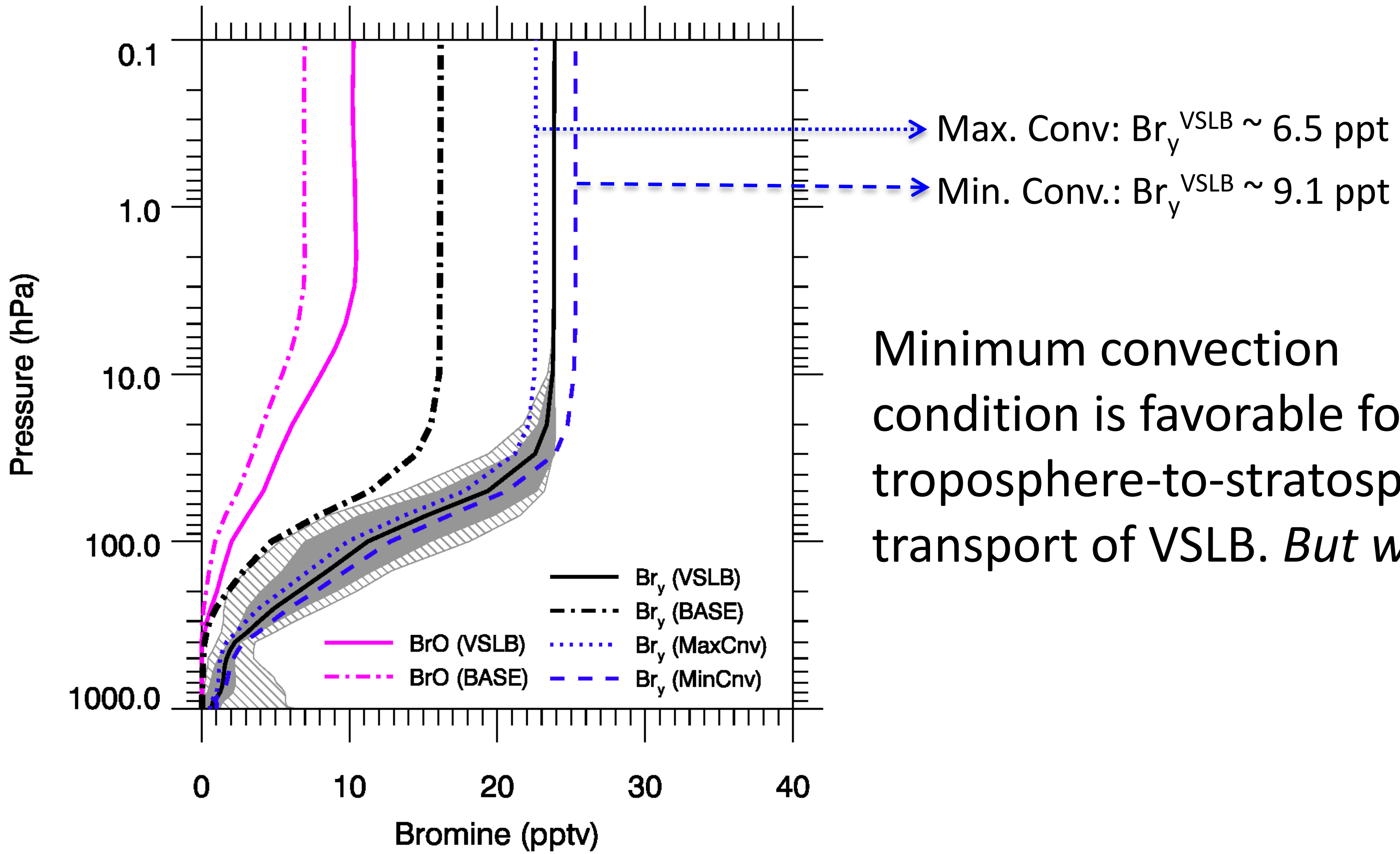
Source gas and product gas injection through TTL



Above 355K, > half of VSLB is converted to inorganic products.

➔ *If one needs to quantity Br_y^{VSLB} using near-tropopause measurements, need to measure both source gases and product gases.*

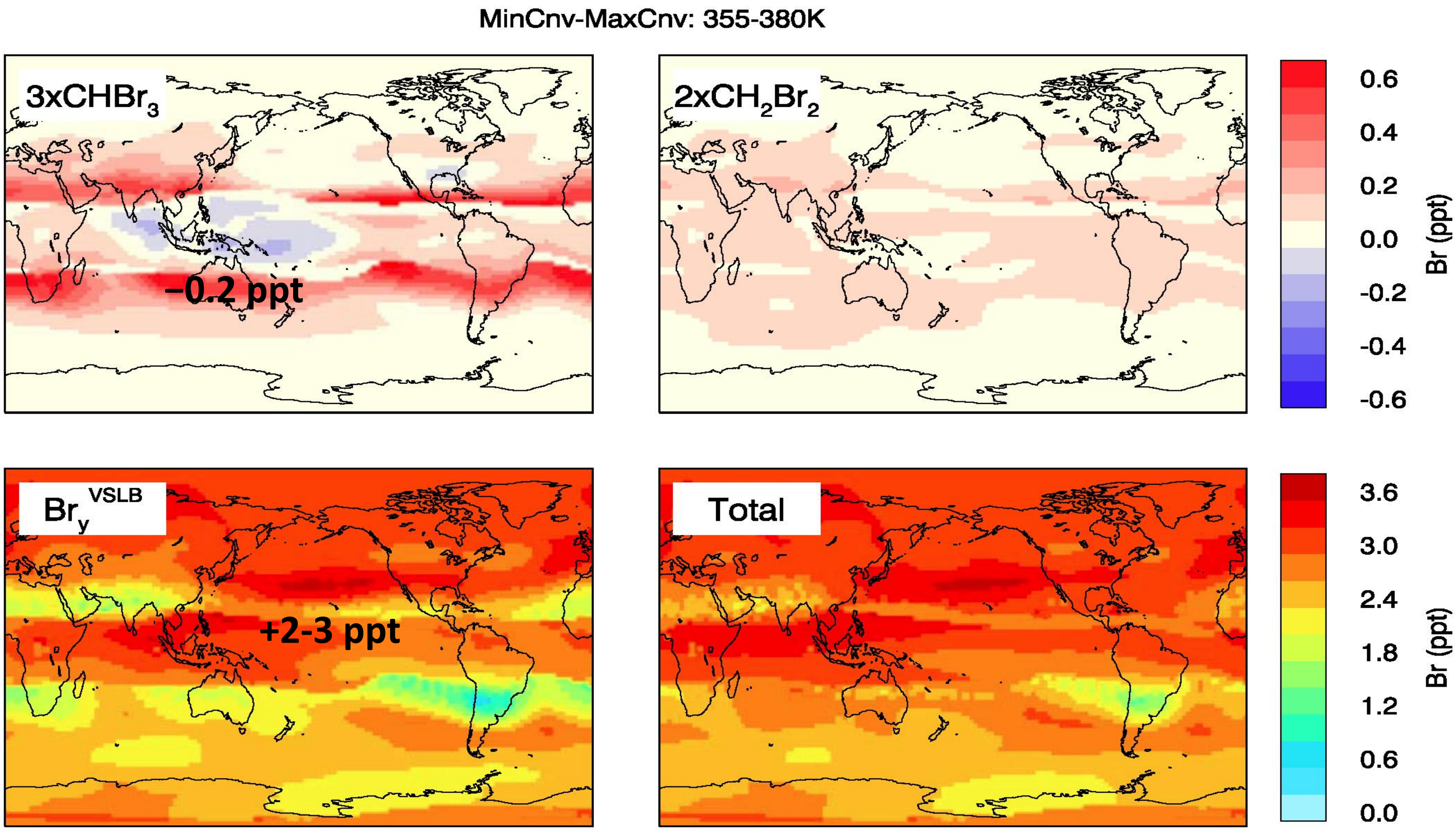
Sensitivity of Br_y^{VSLB} to convection strength



Minimum convection condition is favorable for troposphere-to-stratosphere transport of VSLB. *But why?*

Minimum vs. Maximum convection

10-year averaged differences in $CHBr_3$, CH_2Br_2, Br_y^{VSLB} between minimum and maximum convection simulations



Conclusions

- Bromoform ($CHBr_3$) and dibromomethane (CH_2Br_2), contribute ~7.7 pptv to the present-day stratospheric inorganic bromine.

  (Likely an overestimate (Hossaini et al.,2013), need more tropical surface measurements to constrain emissions.

- The most active transport of VSLB from the marine boundary layer through the tropopause occurs over (i) the tropical Indian Ocean, (ii) the western Pacific warm pool, and (iii) off the Pacific coast of Mexico.

- Surface concentrations of VSLB in these critical regions set how much Br_y^{VLSB} can enter the stratosphere. More than half of the VSLB enter lower stratosphere through TTL product gas injection.

  (Are these model predictions true in real atmosphere?)

- Differences in the strength of deep convection can introduce a ~2.6 ppt uncertainty in Br_y^{VSLB}, with minimum convection condition more favorable for VSLB TST.

Wishlist

- Near-surface measurements of VSL bromocarbons in the tropics:

  i) to constrain emissions;

  ii) the 1st needed component to test the hypothesis "whatever presents in the near-surface enters the TTL";

  iii) potential future missions targeting the tropical Indian Ocean.

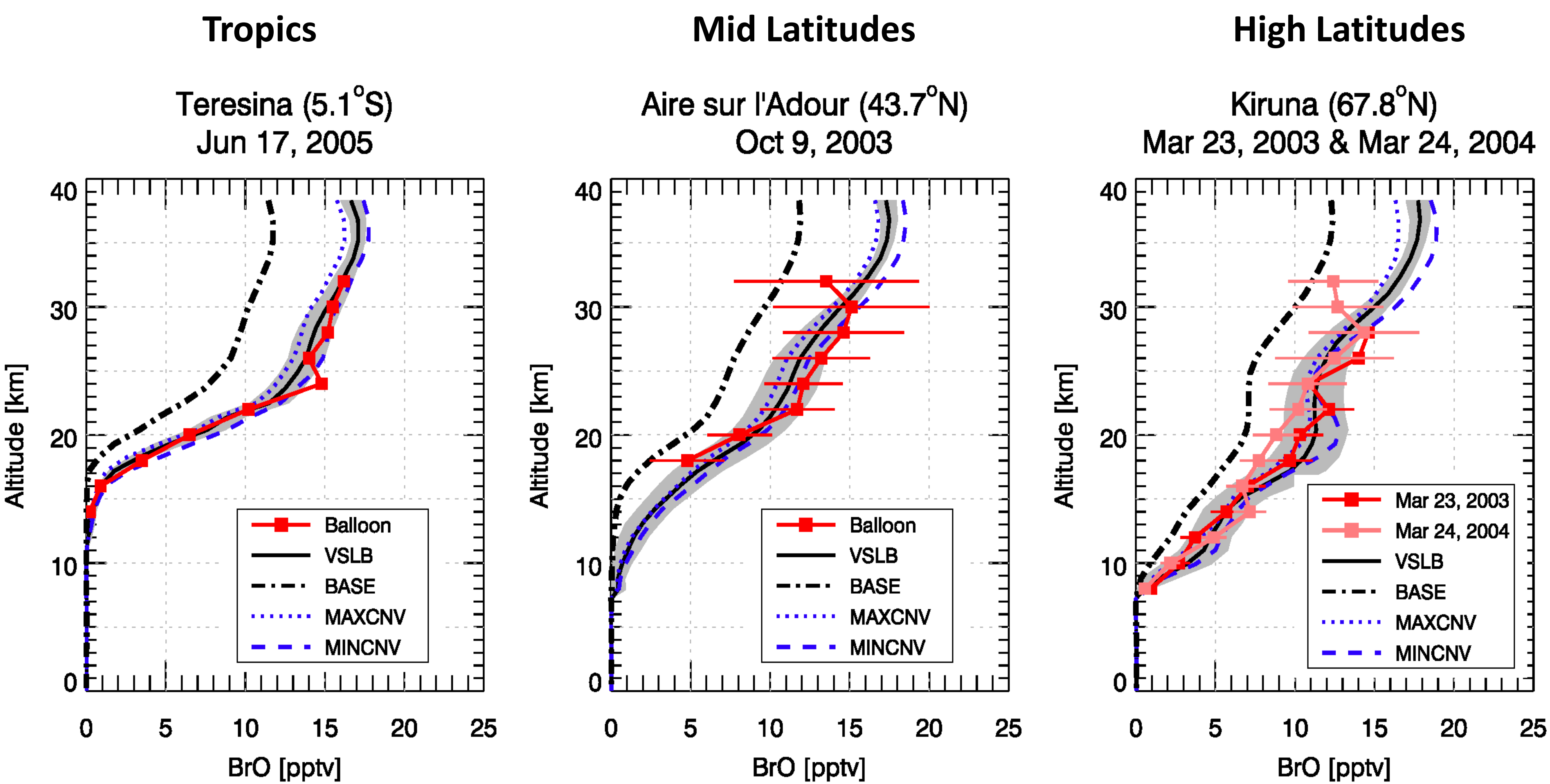
- Measurements of the full composition of organic + inorganic bromine in the TTL (100-150hPa):

  i) the 2nd needed component to test the hypothesis "whatever presents in the near-surface enters the TTL";

  ii) to quantify source gas injection vs. product gas injection

  iii) to quantify the rainout/washout efficiency of Br_y through the TTL.

Modeled BrO vs. Balloon Observations



. (*However, BrO measurement alone seems inadequate to constrain Br_y^{VSLB} to the precision we need.*)