Institute for Climate and Atmospheric Science
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Growth in stratospheric loading of very short-lived substances and their impact on ozone and climate

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Boulder
Tuesday 21st July 2015

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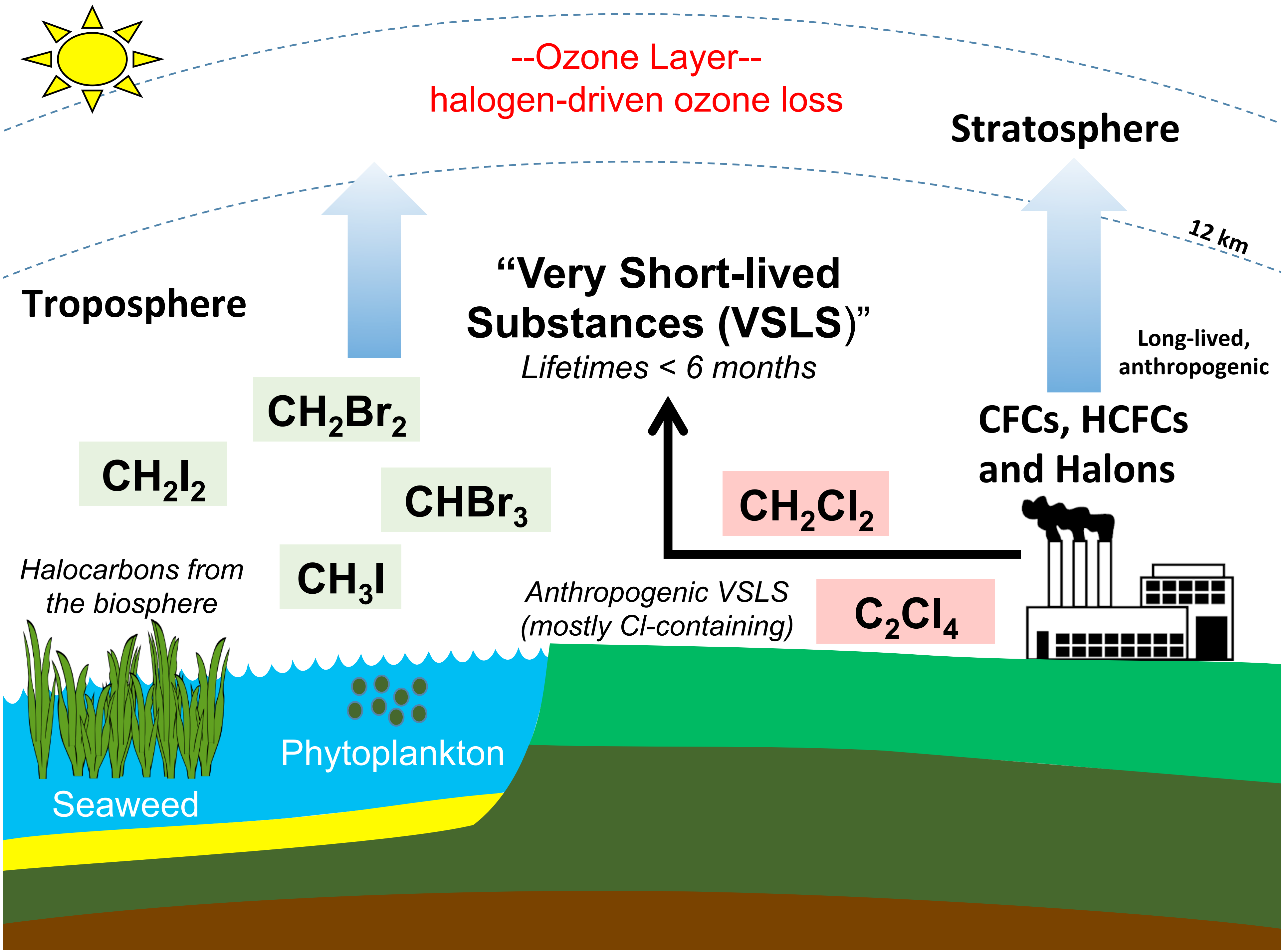


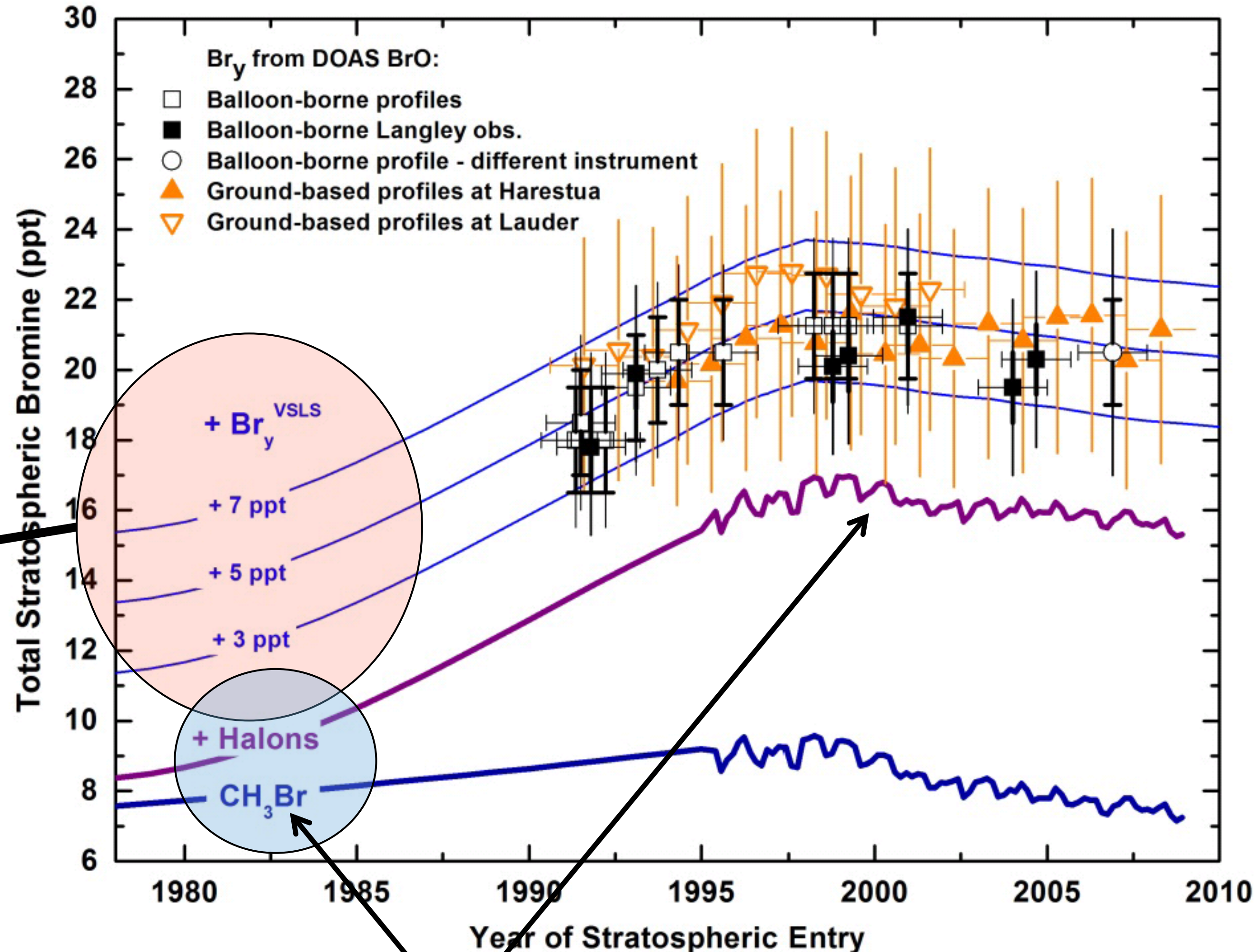
--Ozone Layer--
halogen-driven ozone loss
Stratosphere
12 km
Troposphere
"Very Short-lived Substances (VSLS)"
Lifetimes < 6 months
Long-lived, anthropogenic
CFCs, HCFCs and Halons
CH_2Br_2
CH_2I_2
$CHBr_3$
CH_3I
CH_2Cl_2
C_2Cl_4
Halocarbons from the biosphere
Anthropogenic VSLS (mostly Cl-containing)
Phytoplankton
Seaweed

Stratospheric Bromine from VSLS

More bromine in stratosphere that can be explained by long-lived gases alone

Oceanic VSLS estimated to supply 6 (3-8) ppt of bromine in the stratosphere

~20 to 30% of the TOTAL bromine in the stratosphere



Contribution from long-lived gases

M. Dorf, K. Pfeilsticker et al. in WMO (2014)

Key Questions

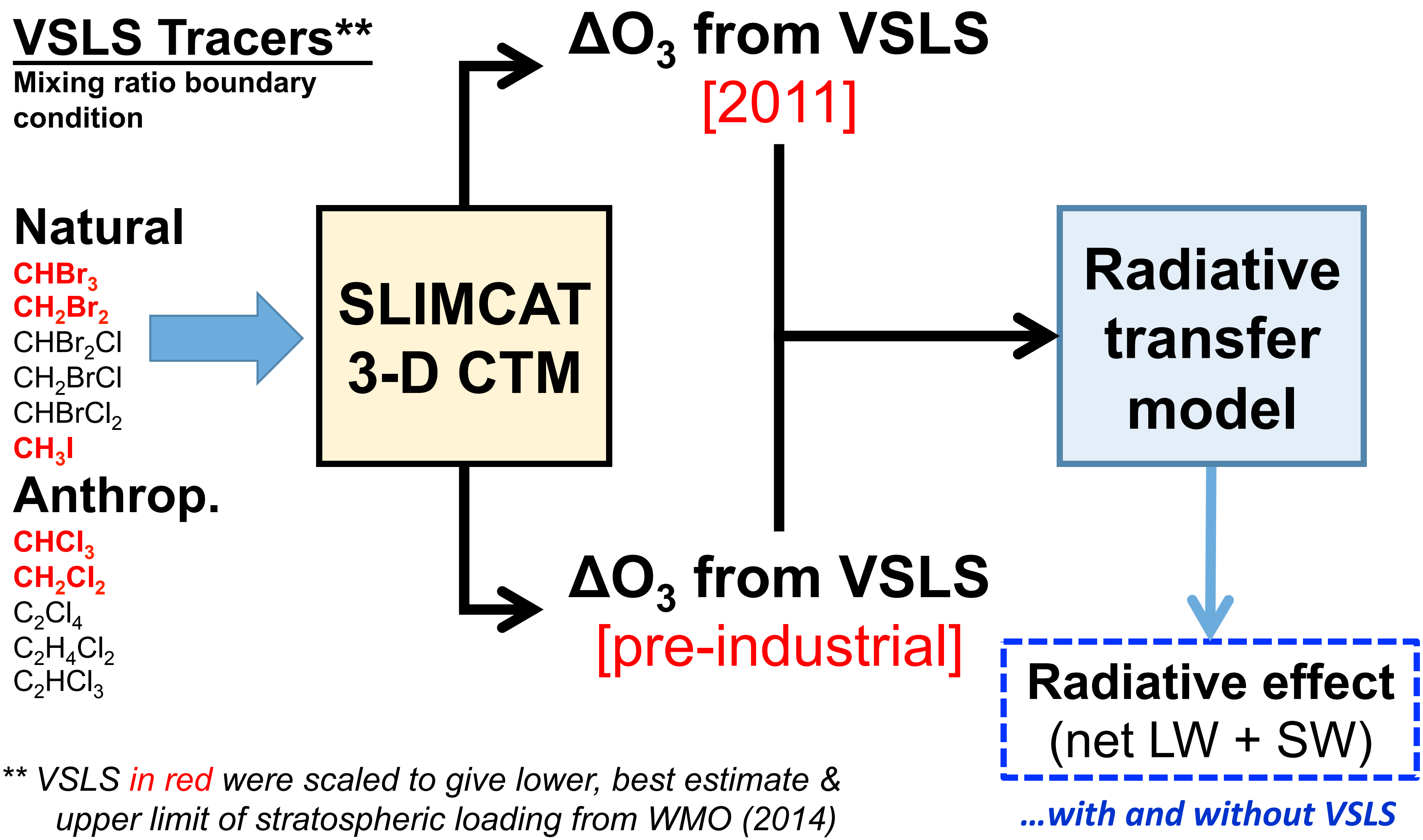
1. **What is the impact of VSLS on ozone in the lower stratosphere?**
   → Which VSLS (natural vs anthropogenic) influence ozone most?

2. **What are the radiative implications?**

3. **Has VSLS-driven ozone loss changed over time?**
   → Contribution to the radiative forcing of strat. ozone

4. **Is the stratospheric loading of anthropogenic VSLS changing?**

Global Model Simulations

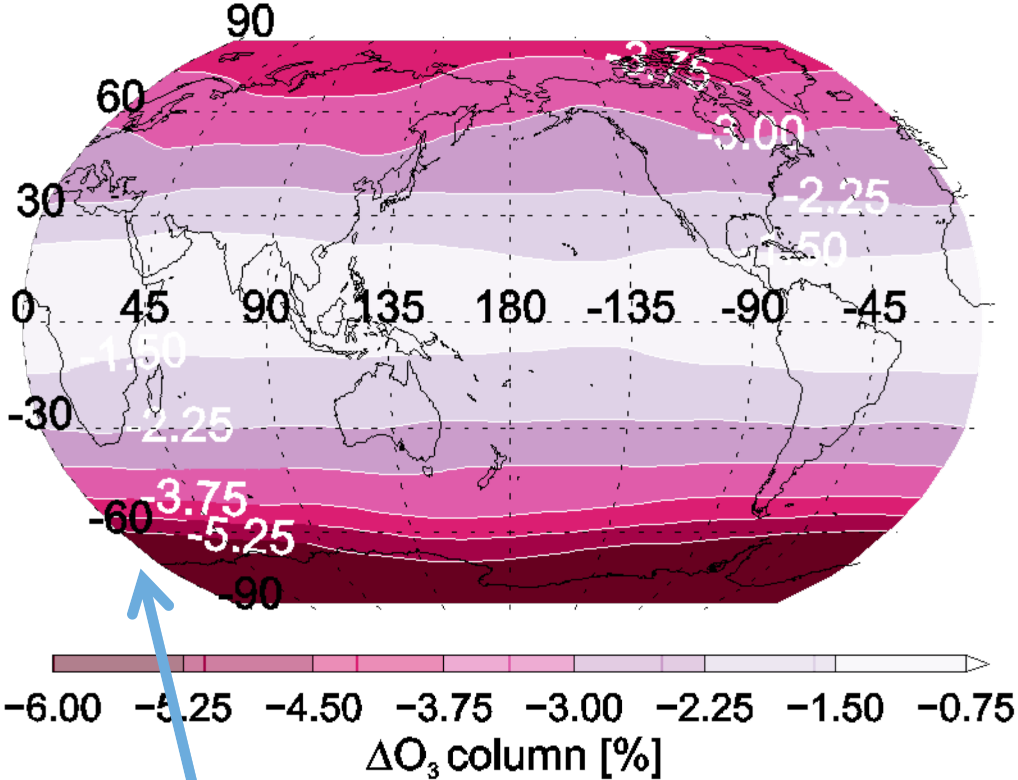


*** VSLS in red were scaled to give lower, best estimate & upper limit of stratospheric loading from WMO (2014)*

*** Time-independent Br and I; trends considered for chlorine.*

ΔO_3 column due to VSLS [2011]

*i.e., the presence or absence of **all** VSLS in the 2011 atmosphere*

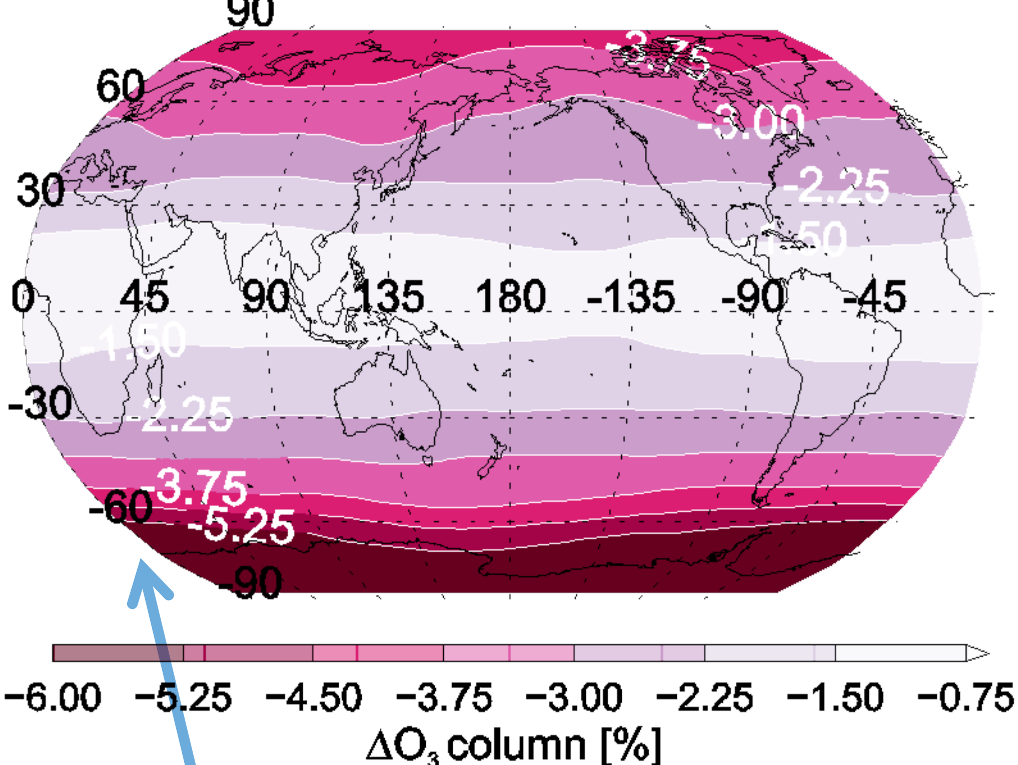


Largest impact at mid to high latitudes

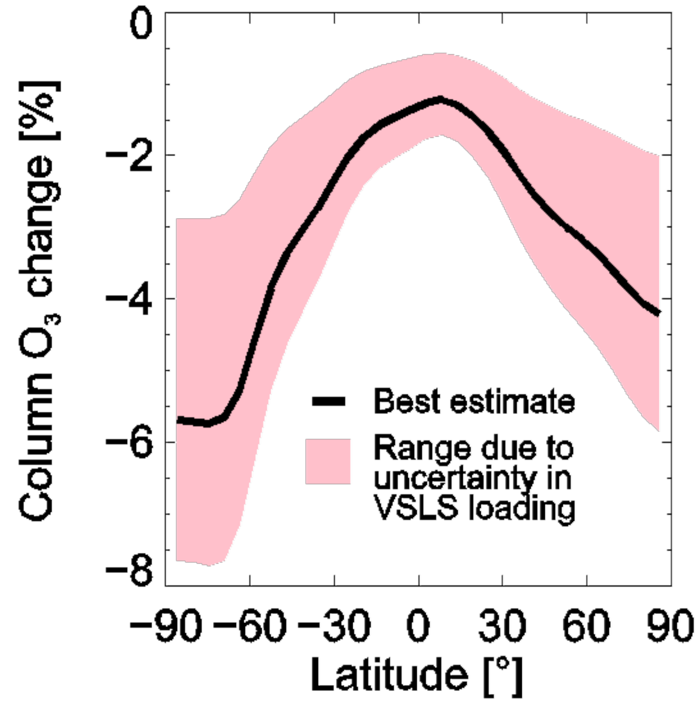
$ClO + BrO$ ---- $Cl + Br + O_2$
$Cl + O_3$ ---- $ClO + O_2$
$Br + O_3$ ---- $BrO + O_2$
Net: $2O_3$ ---- $3O_2$

ΔO_3 column due to VSLS [2011]

*i.e., the presence or absence of **all** VSLS in the 2011 atmosphere*



Uncertainty due to poor constraint on stratospheric loading of VSLS



Largest impact at mid to high latitudes

VSLS reduce O_3 column up to ~6% or ~15 Dobson Units

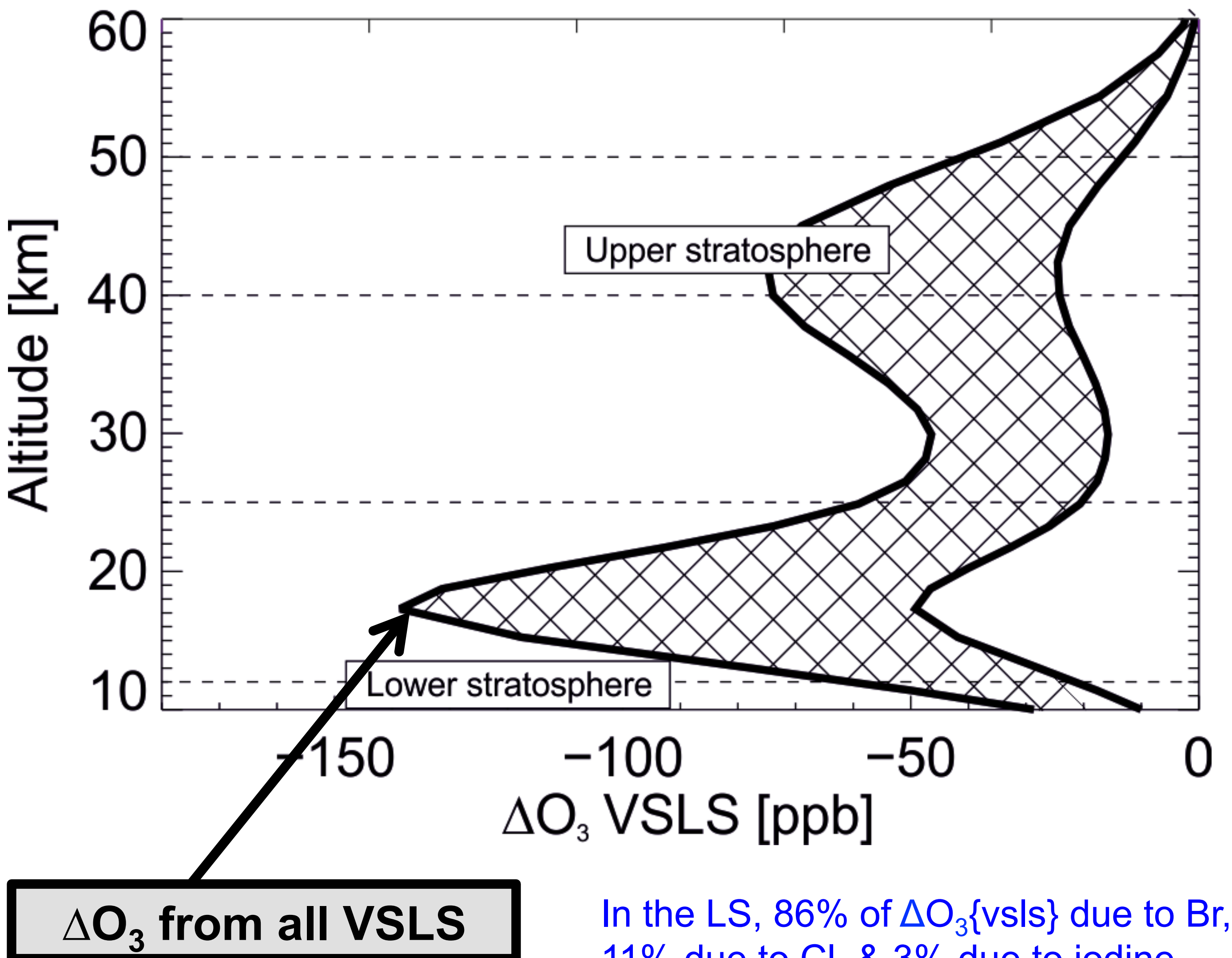
$ClO + BrO$ ---- $Cl + Br + O_2$

$Cl + O_3$ ---- $ClO + O_2$

$Br + O_3$ ---- $BrO + O_2$

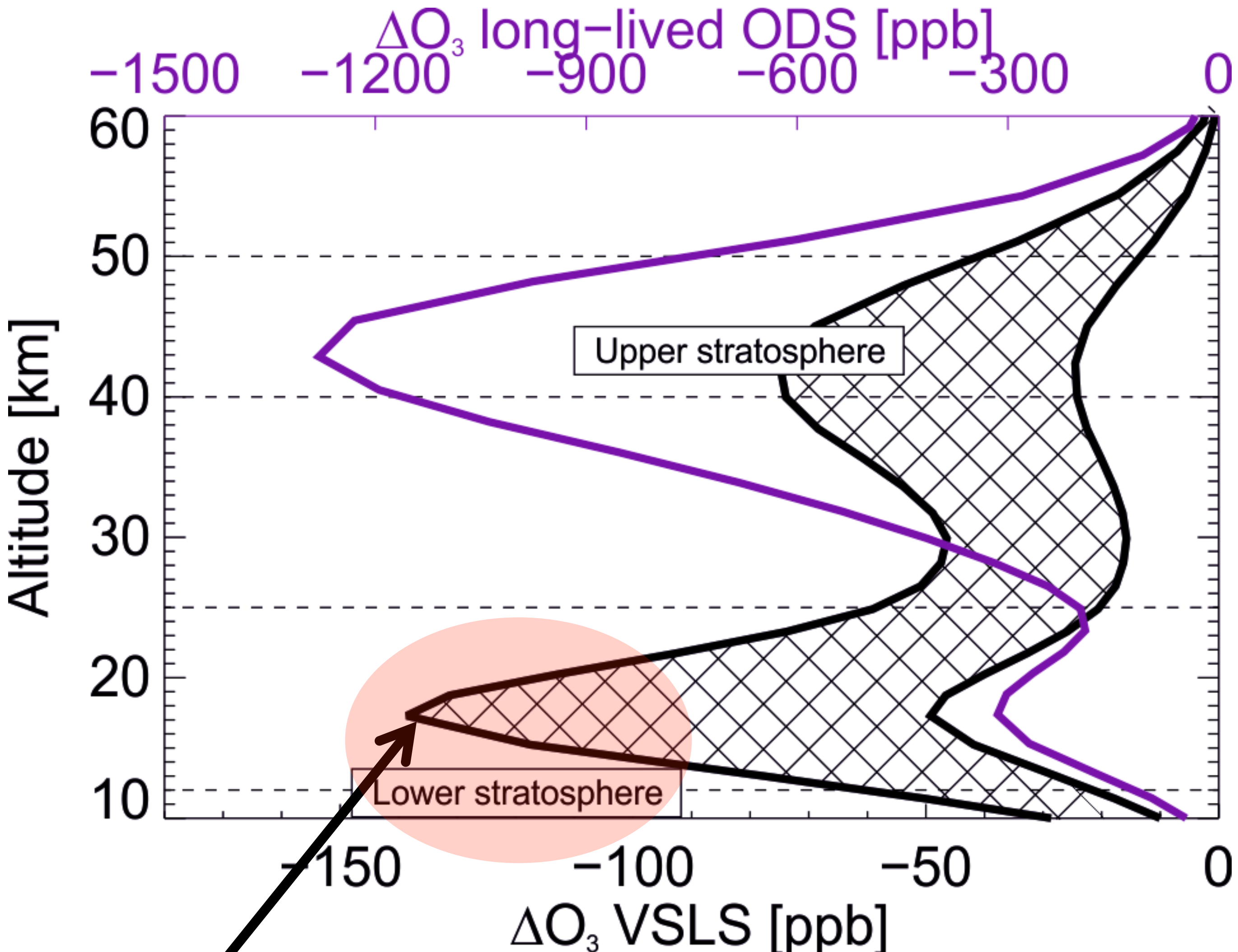
Net: $2O_3$ ---- $3O_2$

Altitude-resolved ΔO_3 due to VSLS



In the LS, 86% of ΔO_3\{vsls\} due to Br, 11% due to Cl, & 3% due to iodine.

Altitude-resolved ΔO_3 due to VSLS



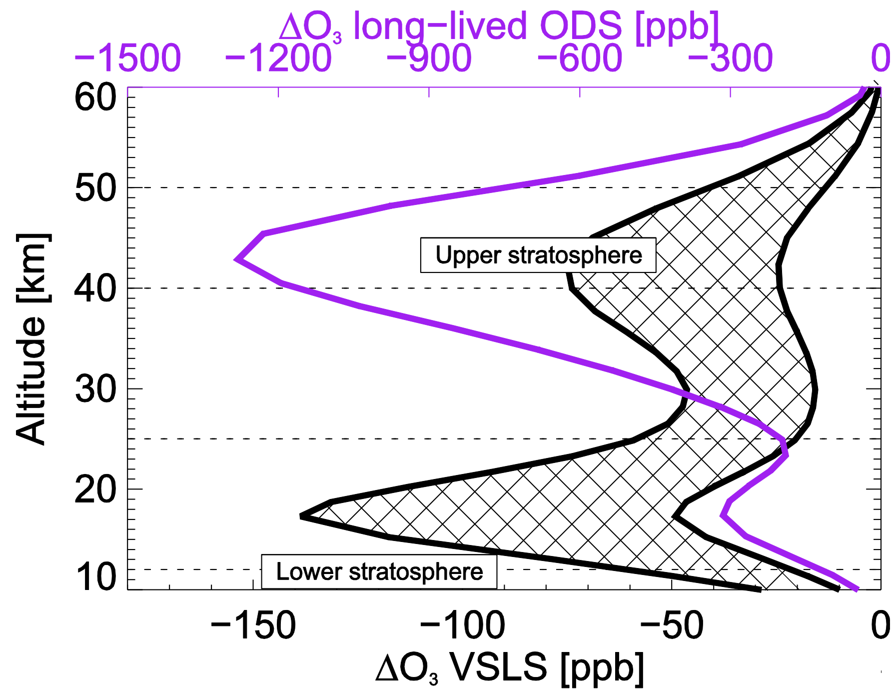
ΔO_3 from long-lived gases (CFCs etc.)

Per molecule, O_3 perturbations in UTLS cause larger radiative effects

ΔO_3 from all VSLS

In the LS, 86% of ΔO_3\{vsls\} due to Br, 11% due to Cl, & 3% due to iodine.

Radiative Effect (RE) of O_3 loss



Long-lived ODSs
RE of –0.17 Wm^{-2}

Caused by >3000 ppt equivalent Cl

Bromine VSLS
RE of –0.07 Wm^{-2}

Caused by (just) several hundred ppt equivalent Cl

VSLS O_3 loss efficient at influencing climate

Normalised, O_3 Radiative Effect due to VSLS is 4x larger than that from long-lived gases

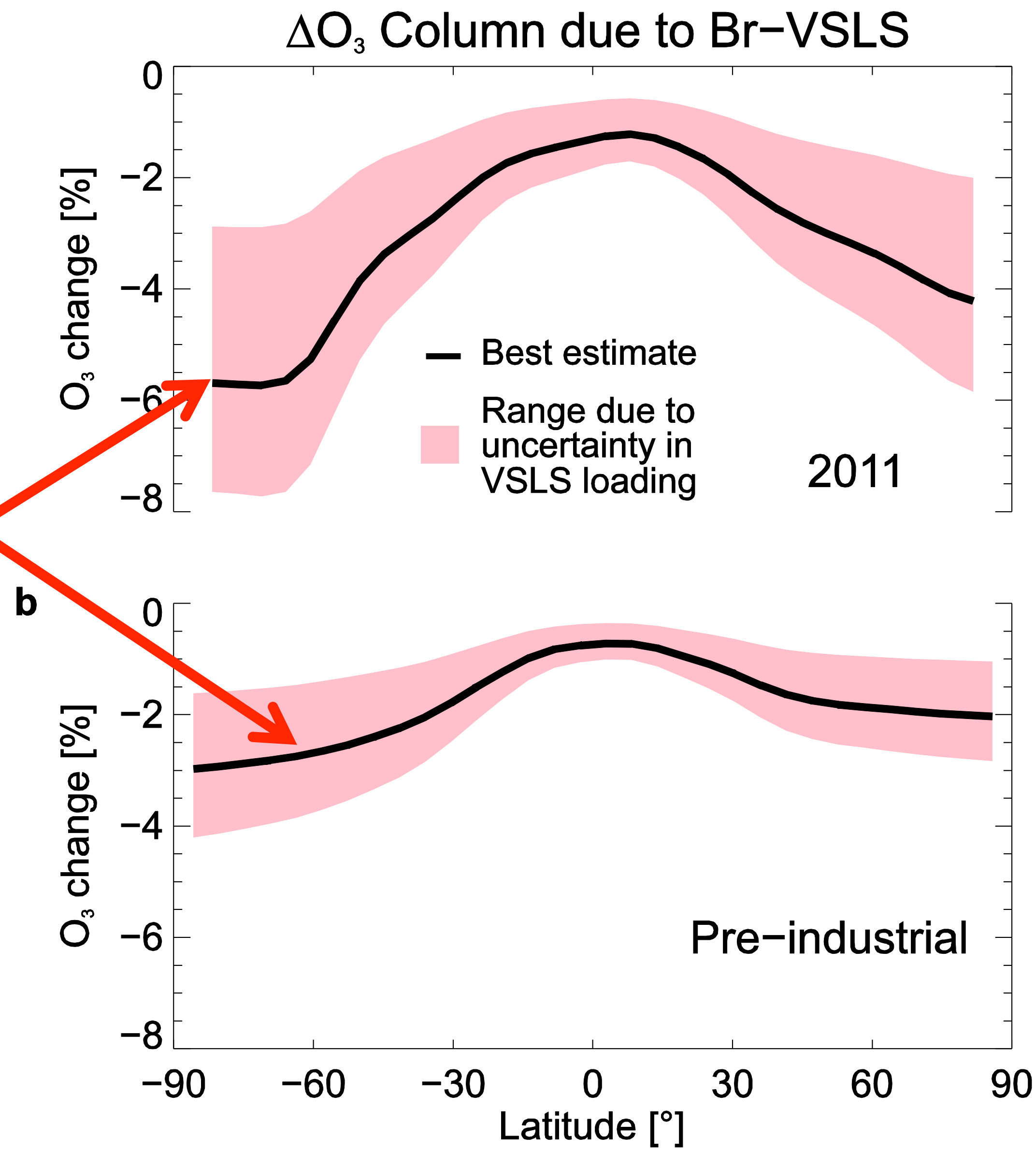
Is there a trend in VSLS-driven O_3 loss?

Yes. For two reasons…

[Reason 1:]

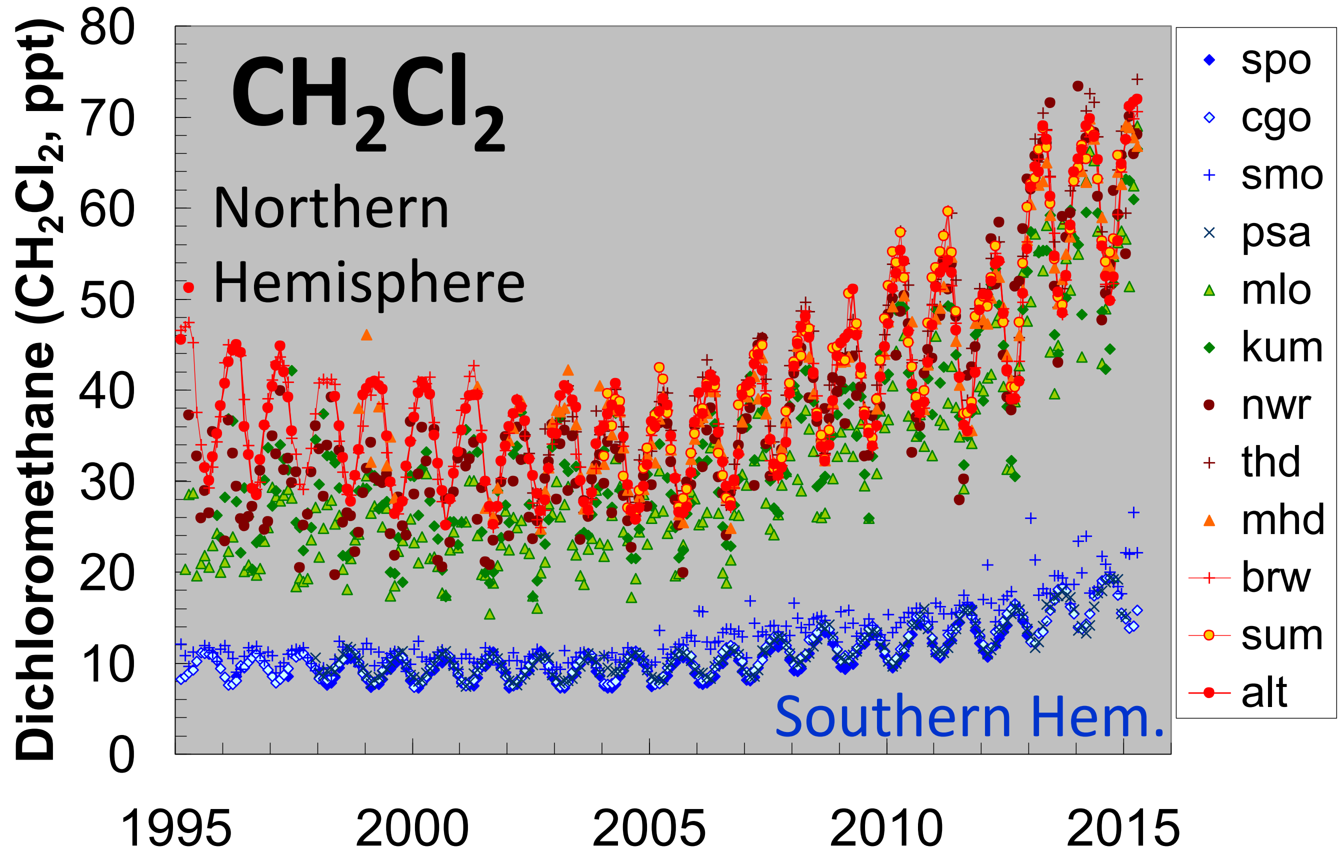
Increase in stratospheric chlorine (from long-lived ODSs) has enhanced *natural* VSLS-driven O_3 loss through coupled Br-Cl cycles

Stratospheric O_3 *Radiative Forcing* calculations should consider VSLS



Trend in VSLS-driven O_3 loss?

[Reason 2.] Some anthropogenic VSLS are increasing throughout the global atmosphere:



7.7%/yr mean increase since 2005

Overall, estimated contribution of <u>-0.02±0.01 Wm^{-2}</u> from VSLS (Cl and Br) to stratospheric O_3 RF since pre-industrial
(bromine accounts for 75% of this RF)

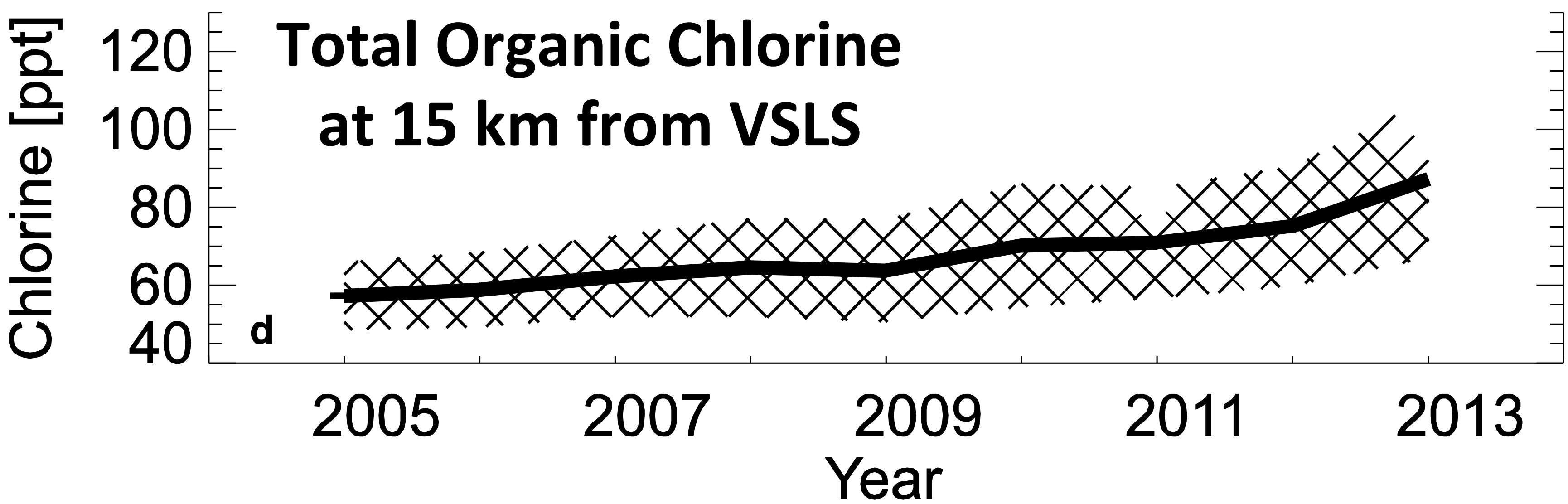
Recent growth in strat. Cl-VSLS

TOMCAT tropospheric model—transporting gases to the stratosphere:
Organic chlorine at 15 km, 20°N to 20°S latitude

Sum Cl = $(2\times CH_2Cl_2) + (3\times CHCl_3) + (2\times C_2Cl_4)$

Model at 15 km: 87 ppt Cl (2013)

Model uses surface observations of CH_2Cl_2, and C_2Cl_4 (NOAA) and $CHCl_3$ (AGAGE) as boundary condition.



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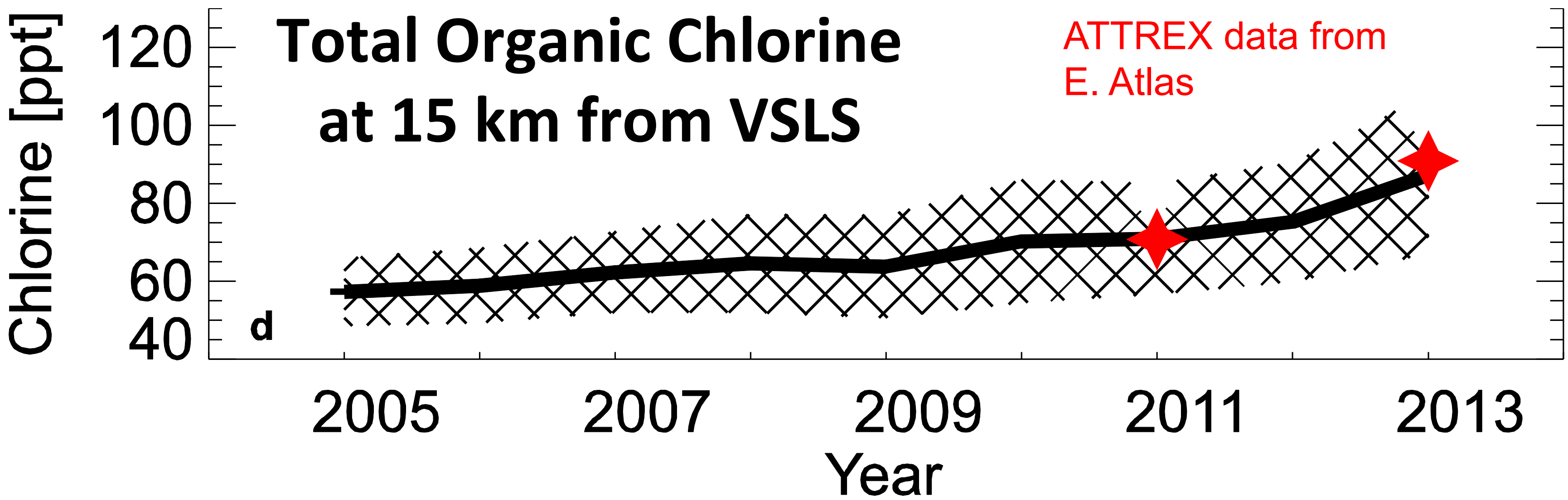
Sum Cl = $(2\times CH_2Cl_2) + (3\times CHCl_3) + (2\times C_2Cl_4)$

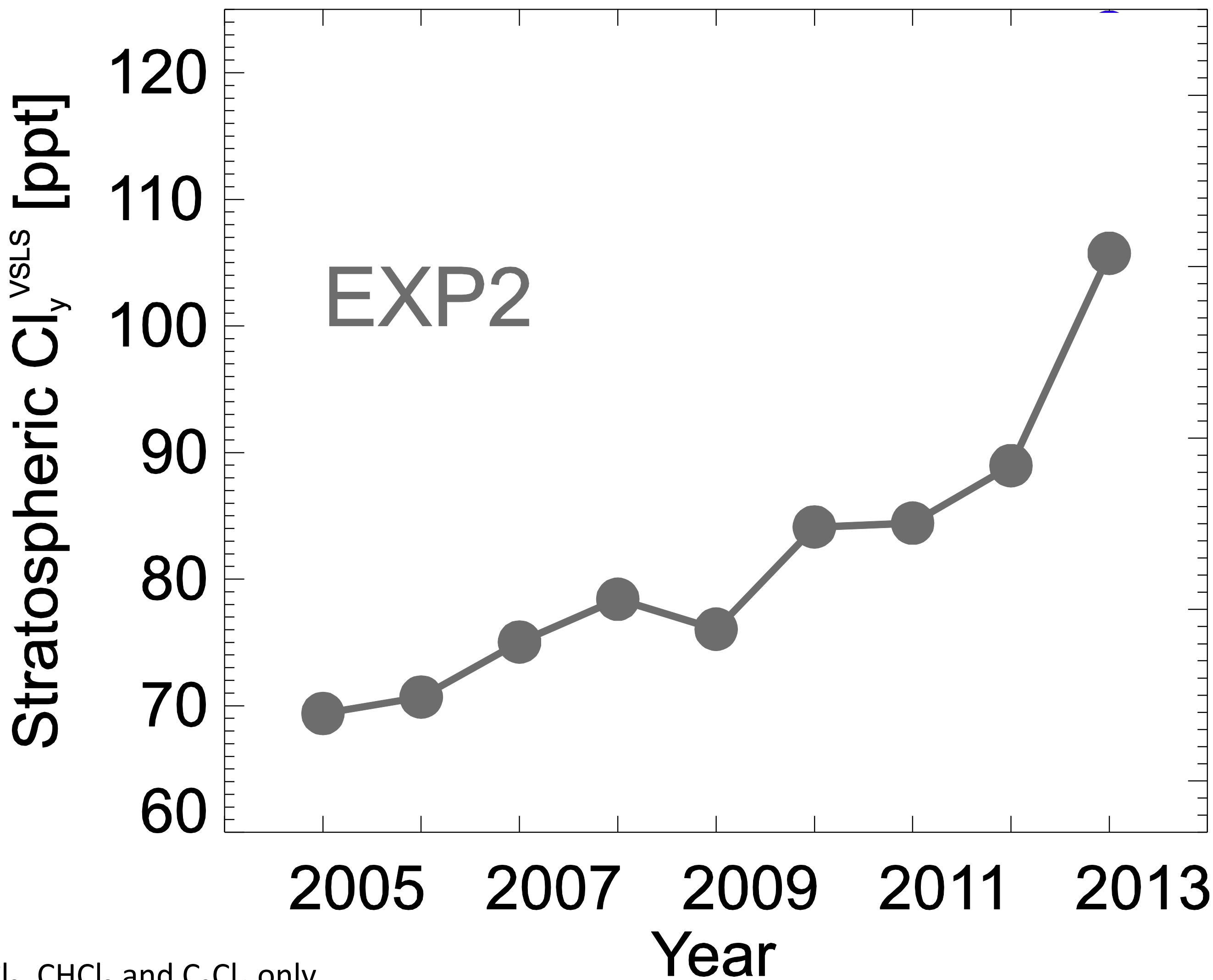
Model at 15 km: 87 ppt Cl (2013)
ATTREX at 15km: 91 ppt Cl (2013)

Model uses surface observations of CH_2Cl_2, and C_2Cl_4 (NOAA) and $CHCl_3$ (AGAGE) as boundary condition.



Modelled trend in Cl_y^{VSLS}

TOMCAT tropospheric model— Including product gases in total Cl amounts



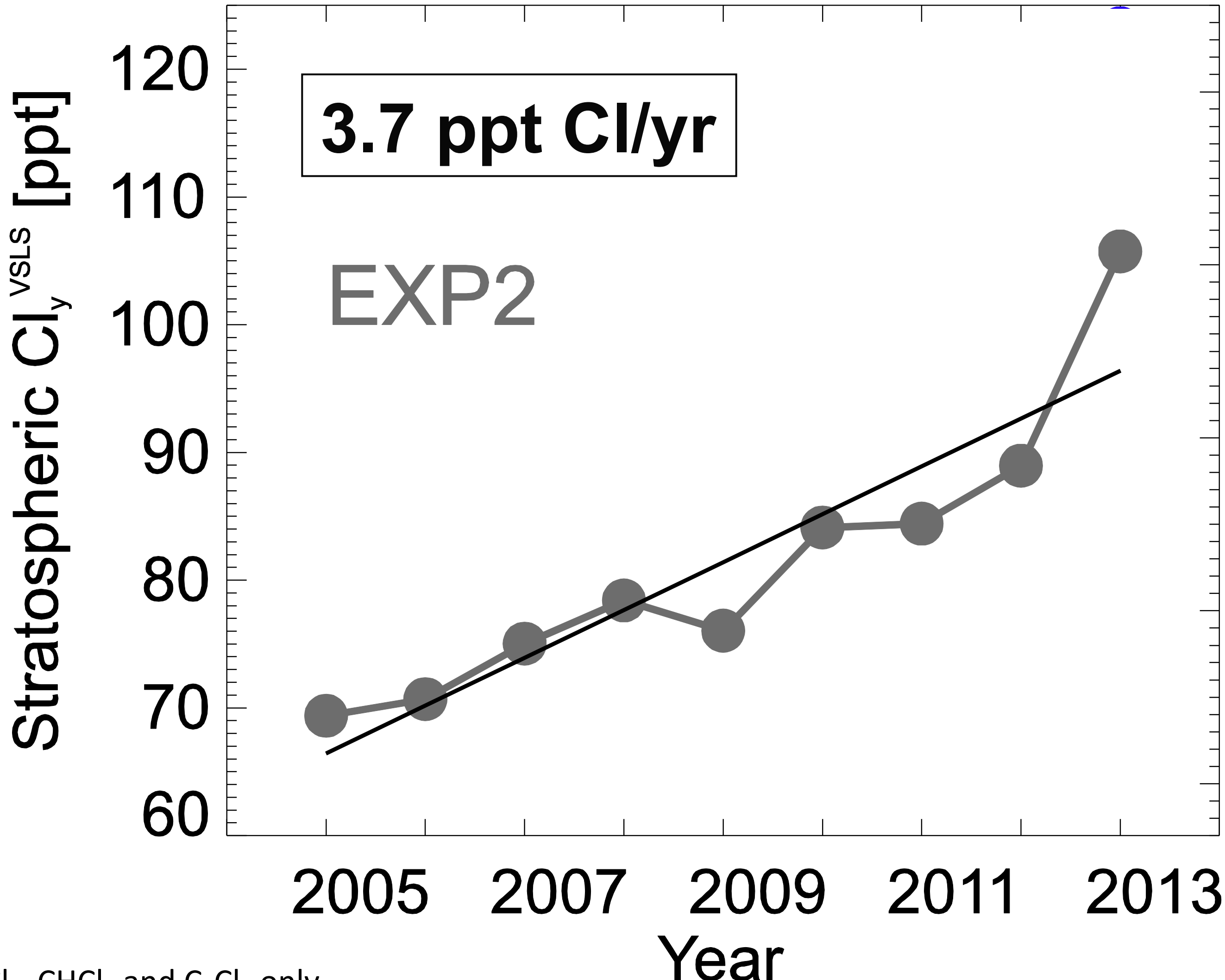
at 15 km in tropics

Product gas injection increases the total by ~18 ppt

EXP2: CH_2Cl_2, $CHCl_3$ and C_2Cl_4 only
EXP3: Sensitivity inc. $C_2H_4Cl_2$ and C_2HCl_3 in addition

Modelled trend in Cl_y^{VSLS}

TOMCAT tropospheric model— Including product gases in total Cl amounts



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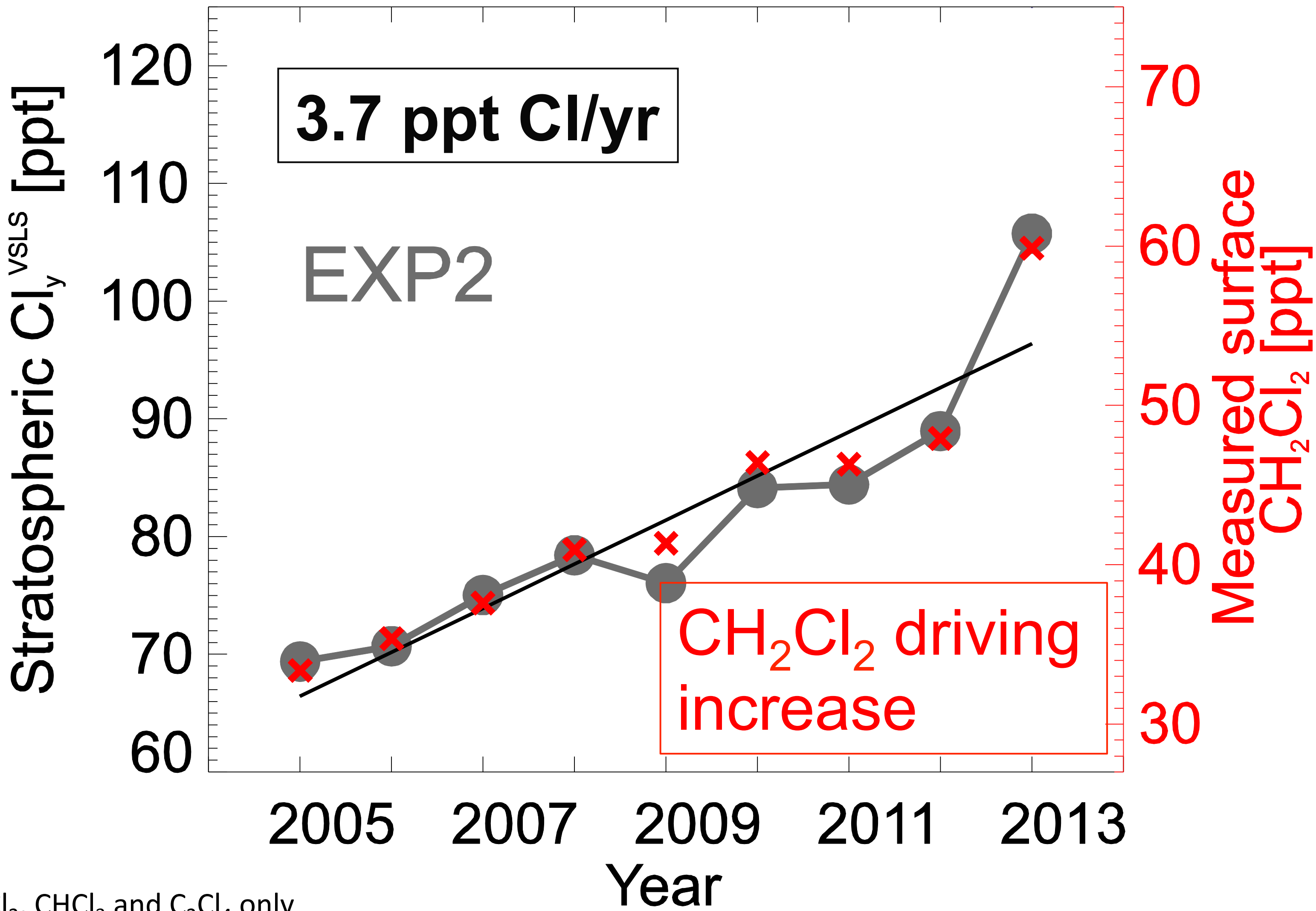


EXP2: CH_2Cl_2, $CHCl_3$ and C_2Cl_4 only

EXP3: Sensitivity inc. $C_2H_4Cl_2$ and C_2HCl_3 in addition

Summary

- **Significant impact of VSLS on LS O_3**
  - → ΔO_3 of ~8-12% (*for an atmosphere without VSLS*)
  - → O_3 change is mostly from natural Br-containing VSLS
- **VSLS-driven O_3 loss is efficient at influencing climate**
  - → Radiative effect of -0.1 Wm^{-2} (stratosphere in 2011)
  - → 4x more efficient than CFCs at influencing climate
  - → Small contribution (-0.02 Wm^{-2}) to strat. O_3 R. Forcing
- **Stratospheric Cl from anthropogenic VSLS increasing**
  - → Growth of 3.7 ppt Cl/yr (2005-2013) (vs. -13.4 ppt/yr from LL ODS)
  - → CH_2Cl_2 not controlled by Montreal Protocol

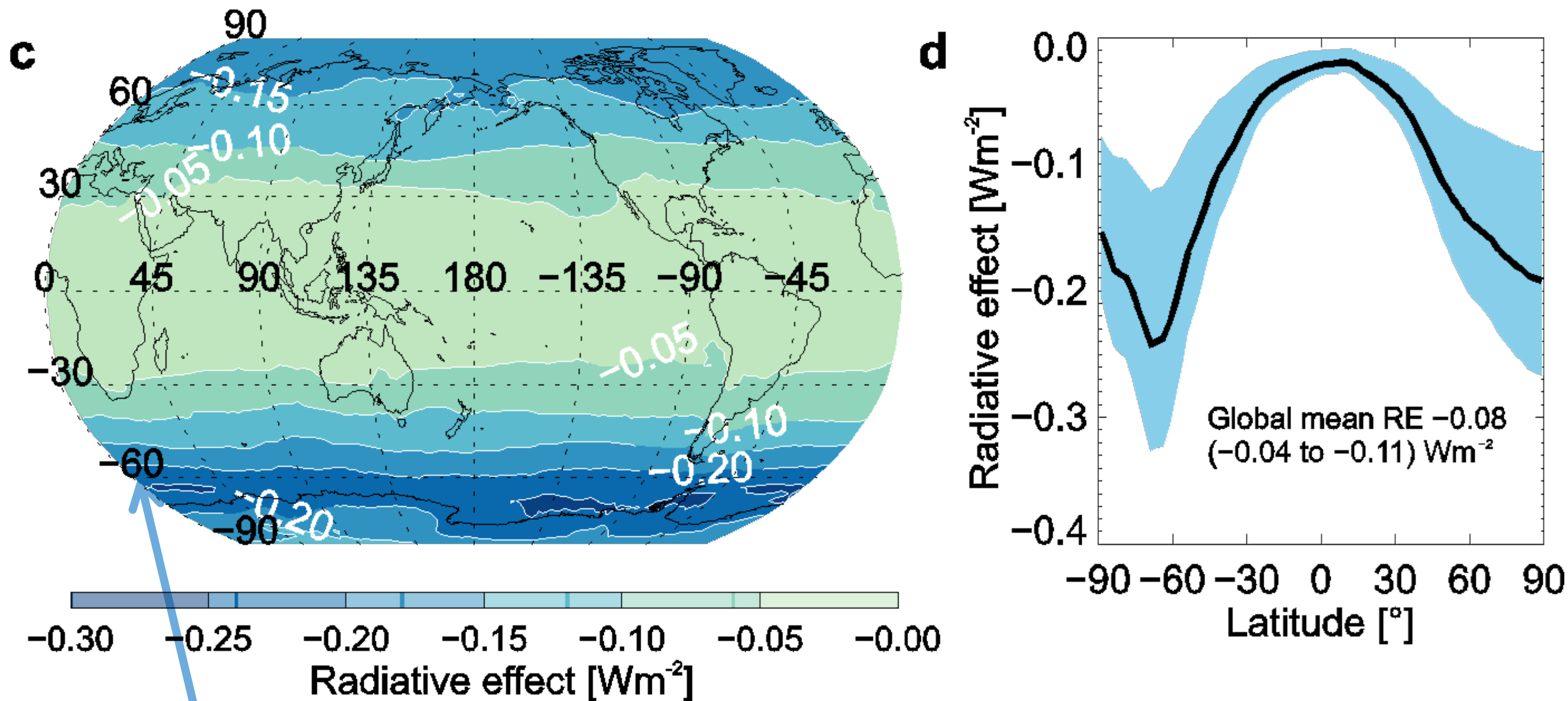
For further details..

Hossaini, R., M.P. Chipperfield, S.A. Montzka, A. Rap, S. Dhomse and W. Feng. (2015a), ***Efficiency of short-lived halogens at influencing climate through depletion of stratospheric ozone***, **Nature Geosciences**, 8, 186-190.

Hossaini, R., M.P. Chipperfield, A. Saiz-Lopez, J. Harrison, R. von Glasow, R. Sommariva, E. Atlas, M. Navarro, S.A. Montzka, W. Feng, S. Dhomse, C. Harth, J. Muhle, C. Lunder, S. O'Doherty, D. Young, S. Reimann, M. Vollmer, P. Krummel, and P. Bernath. (2015b), ***Growth in stratospheric chlorine from short-lived chemicals not controlled by the Montreal Protocol***, **Geophys. Res. Lett**., 42, 4573-4580.

Misc Slides

Radiative effect of VSLS O_3 loss



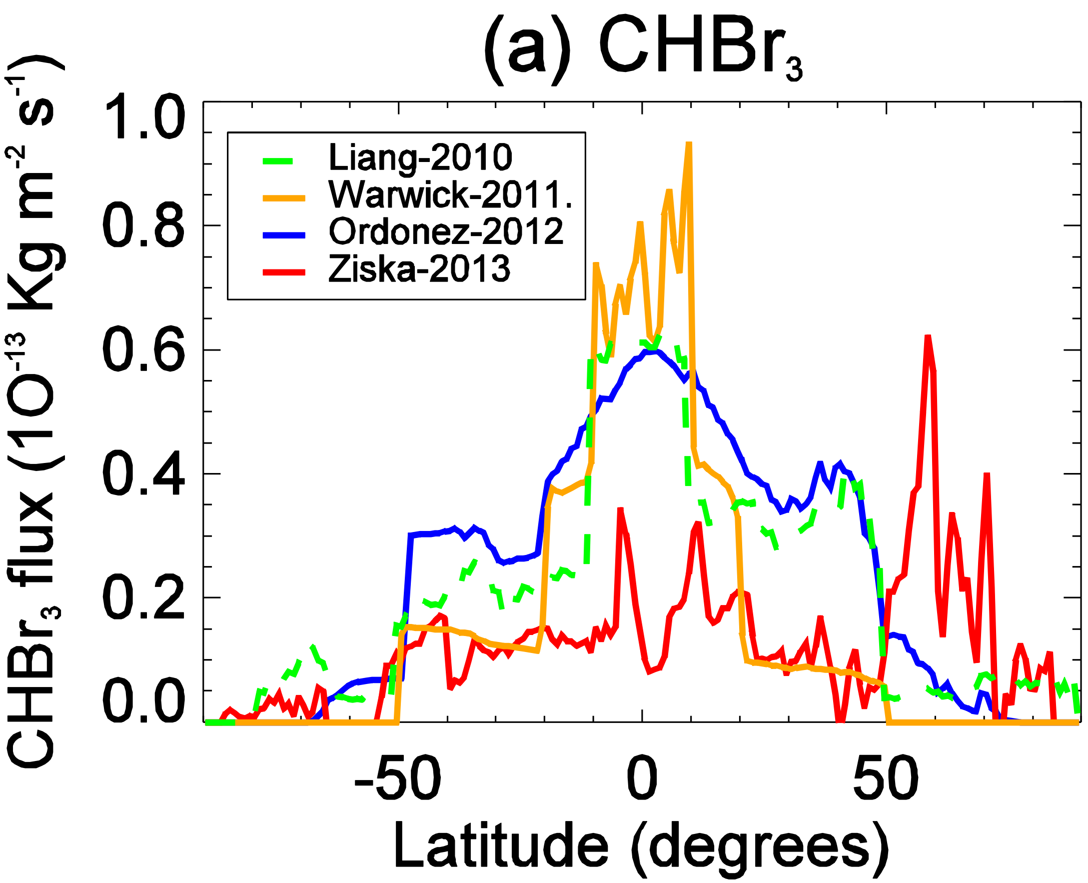
RE of up to -0.30 Wm^{-2}

Global mean RE <u>-0.1 Wm^{-2}</u>

Q. Has VSLS-O_3 loss changed in time?

Radiative forcing?

Uncertainty in $CHBr_3$ emissions

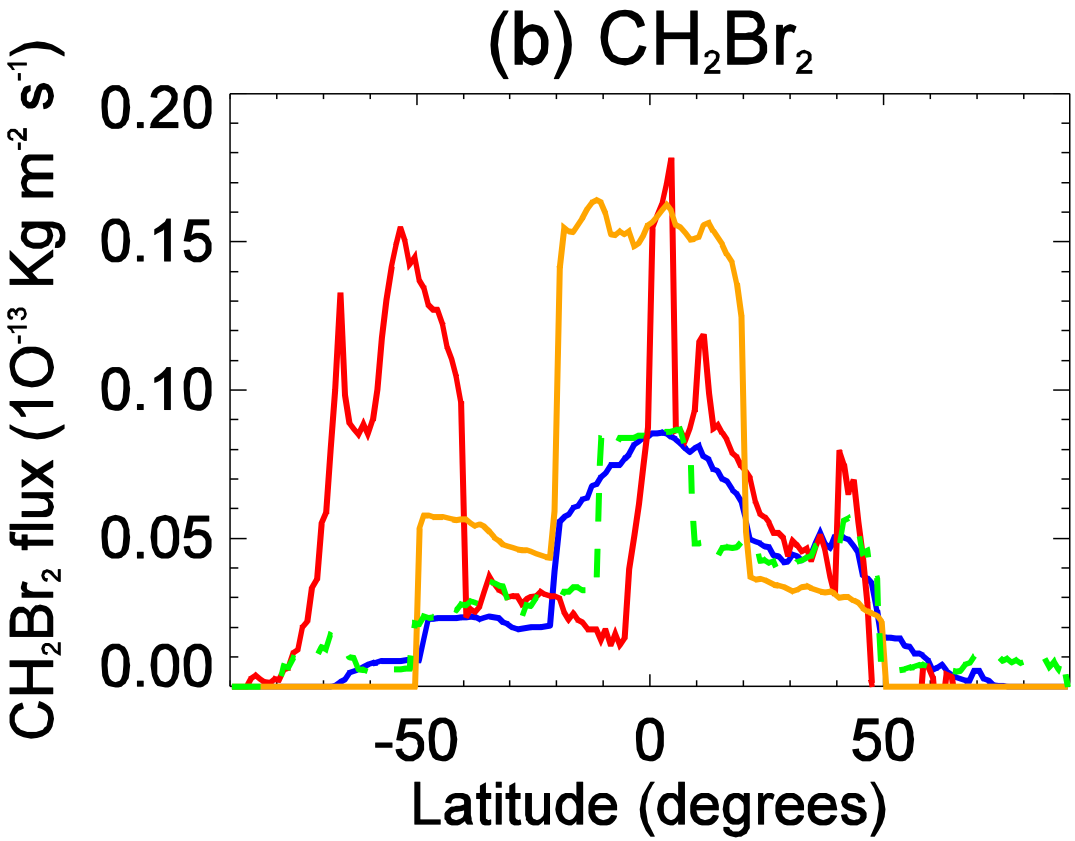


Large uncertainty in distribution and magnitude of emissions

Factor of 3 uncertainty in total global $CHBr_3$ emission

Hossaini et al. (2013, ACP)

Uncertainty in CH_2Br_2 emissions



Large uncertainty in distribution and magnitude of emissions

Factor of 2 uncertainty in total global CH_2Br_2 emission

Hossaini et al. (2013, ACP)

Altitude-resolved ΔO_3 due to VSLS

<u>Natural</u> bromine VSLS are the most important for stratospheric O_3

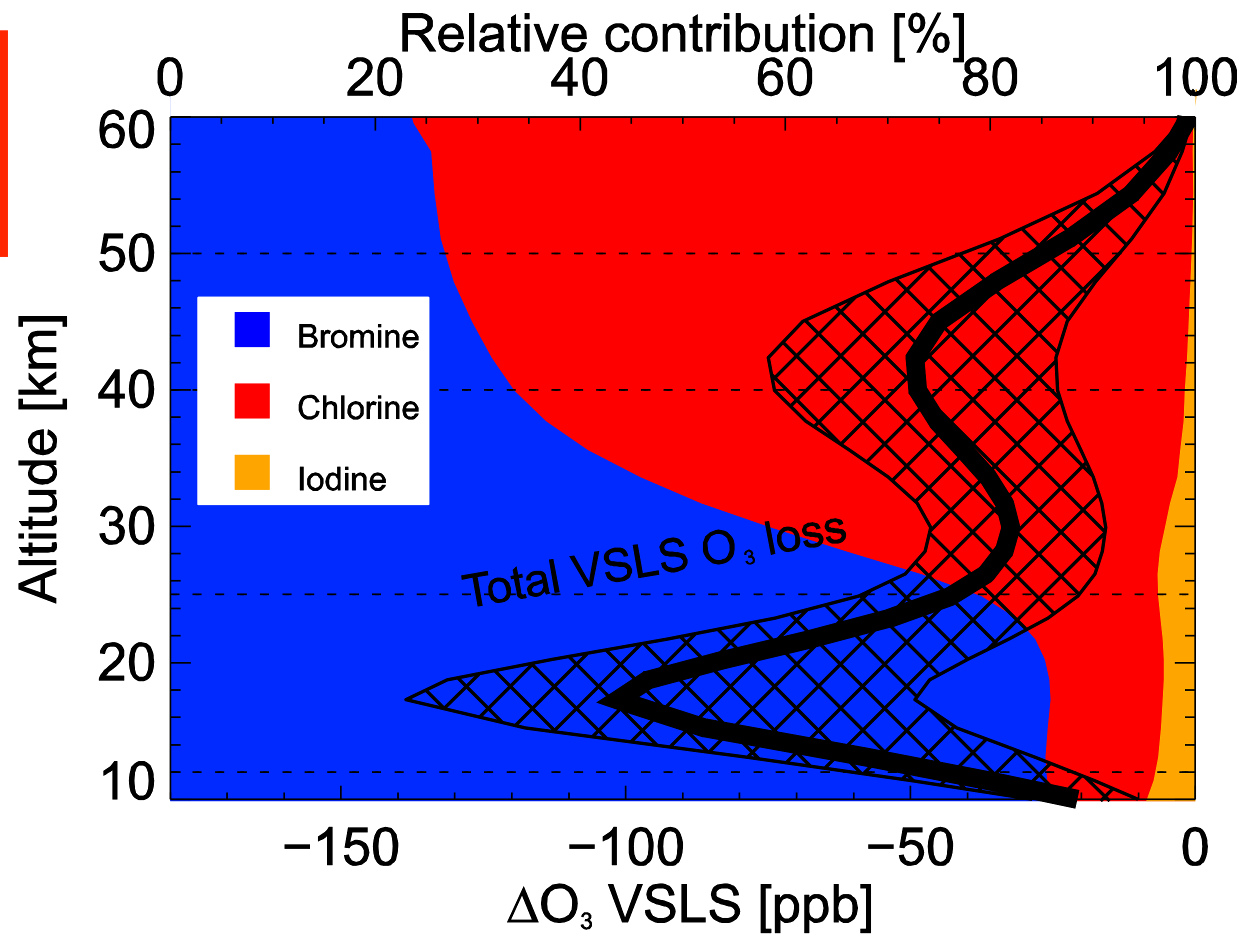
Relative contribution to LS O_3 loss from VSLS

Bromine (86%)

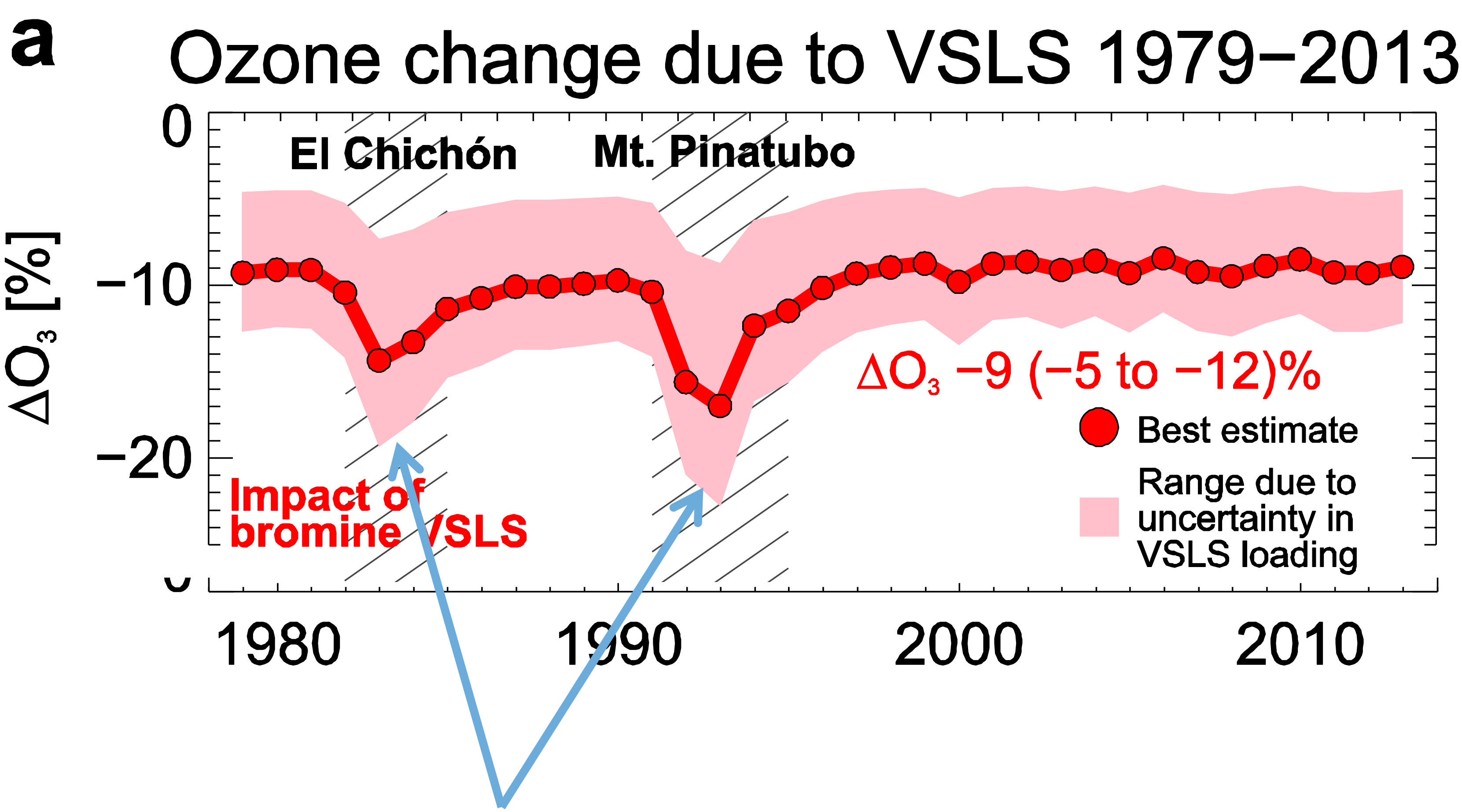
Chlorine (11%)

Iodine (3%)

Chlorine-containing VSLS are of mostly anthropogenic origin

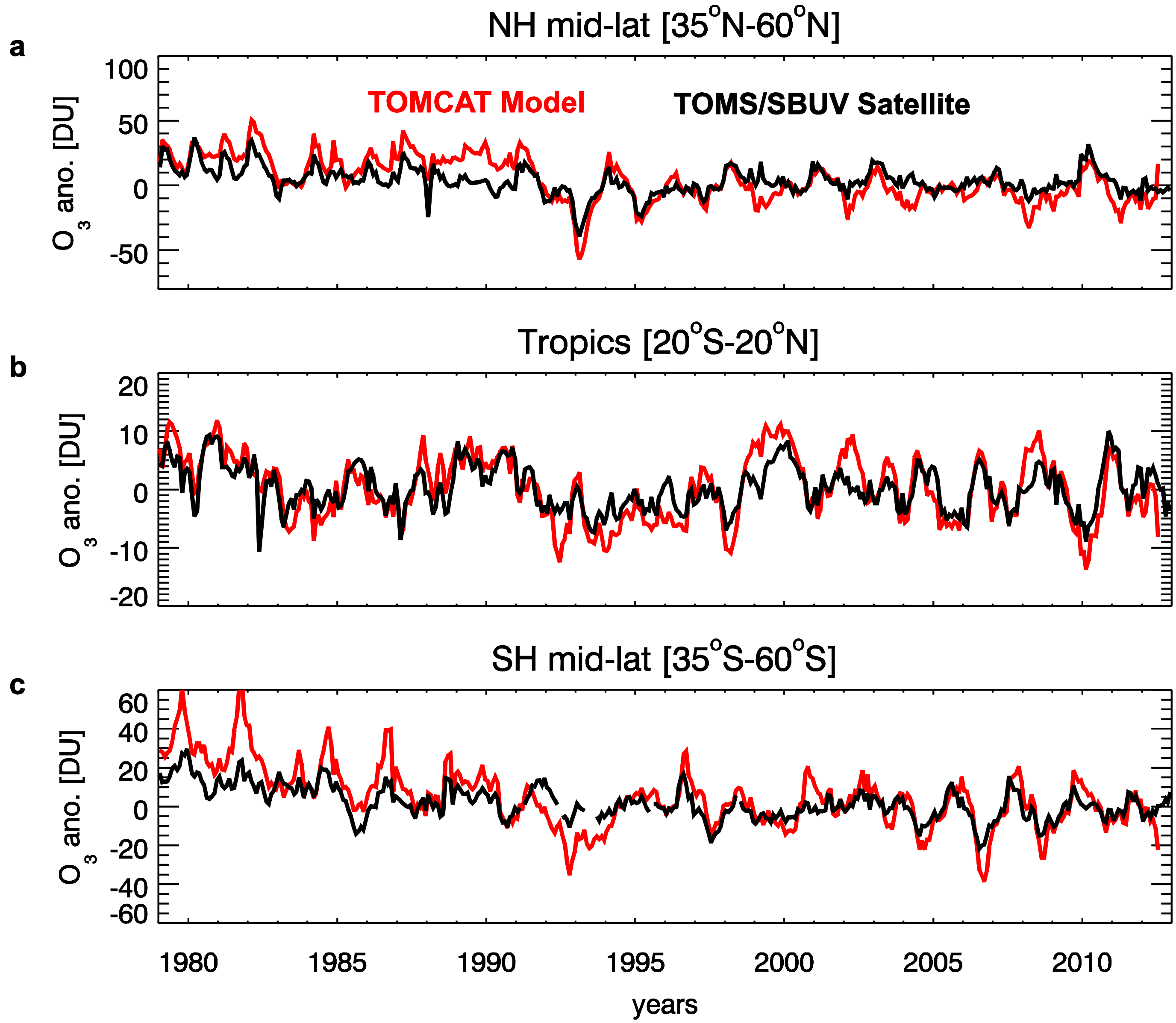


Trend in VSLS-driven O_3 loss?



Enhanced VSLS-driven O_3 loss post volcanic eruptions

[redacted]



NH mid-lat [35°N-60°N]
a
TOMCAT Model
TOMS/SBUV Satellite
O3 ano. [DU]
100
50
0
-50
Tropics [20°S-20°N]
b
20
10
0
-10
-20
SH mid-lat [35°S-60°S]
c
60
40
20
0
-20
-40
-60
1980
1985
1990
1995
2000
2005
2010
years

[redacted]

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New ozone-destroying gases on the rise

Doyle Rice, USA TODAY *12:38 p.m. EST February 16, 2015*

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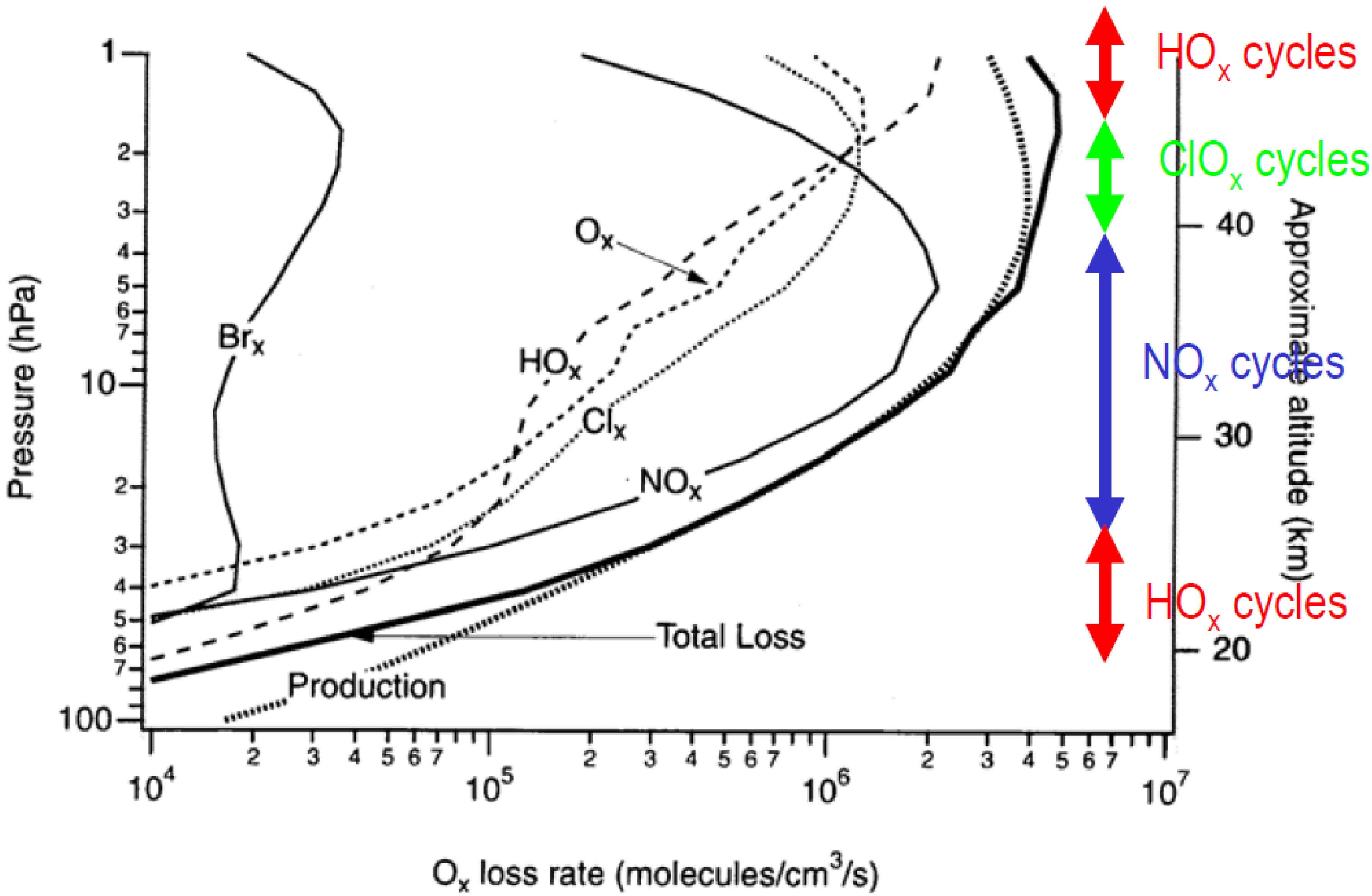
(Photo: AP)

In humanity's ongoing experiment with the Earth's atmosphere, scientists Monday warned of a growing threat from new man-made gases that are chewing away at the ozone layer.

Located high up in the atmosphere, the ozone layer

Vertical distribution of O_x catalytic loss cycles



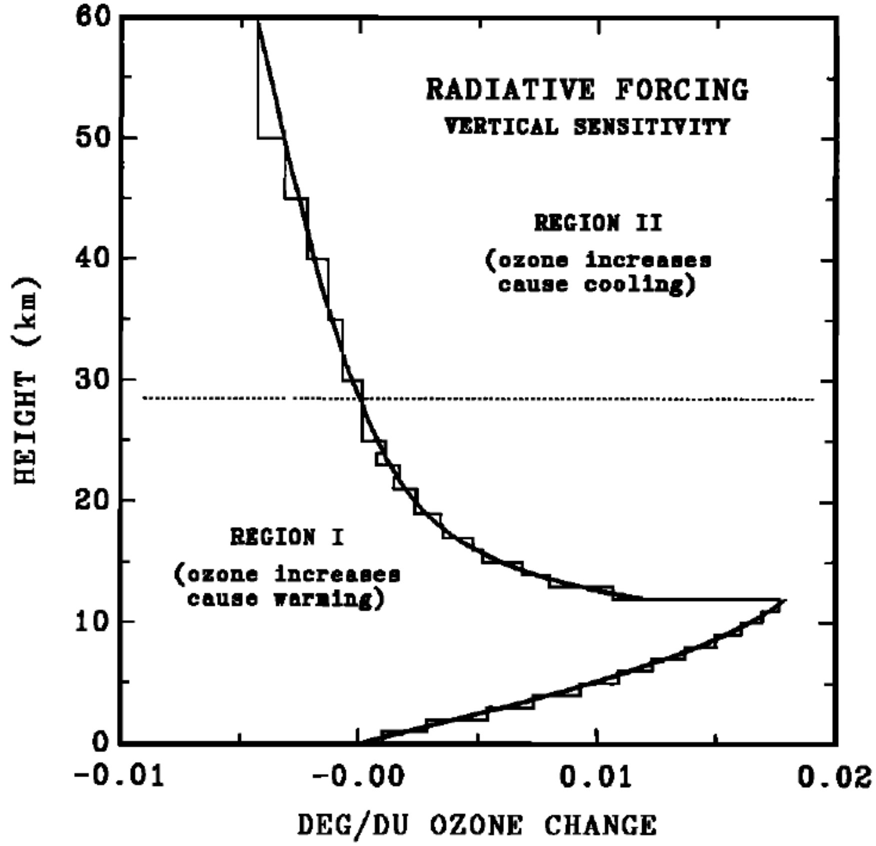


Fig. 1. Radiative forcing sensitivity of global surface temperature to changes in vertical ozone distribution. The heavy solid line is a least squares fit to one-dimensional model radiative-convective equilibrium results computed for 10 Dobson unit ozone increments added to each atmospheric layer. Ozone increases in region I (below ~30 km) and ozone decreases in region II (above ~30 km) warm the surface temperature. No feedback effects are included in the radiative forcing.

Lacis et al. [1990, JGR]

"O_3 increments added near the tropopause produce the largest increase in surface temperature..

Because the greenhouse blanketing produced by a given atmospheric O_3 increment is directly proportional to the temperature contrast between the radiation absorbed and radiation emitted by the O_3 increment; since this temperature contrast is greatest for O_3 increments added near the tropopause, the RF efficiency on a per molecular basis is also greatest for O_3 changes near the tropopause."

Lacis et al. [1990, JGR]