

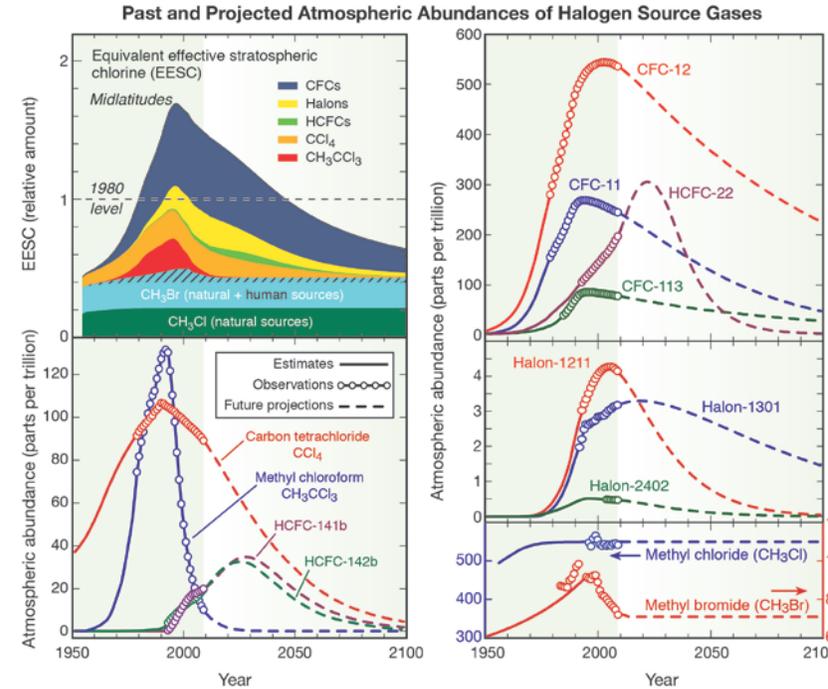
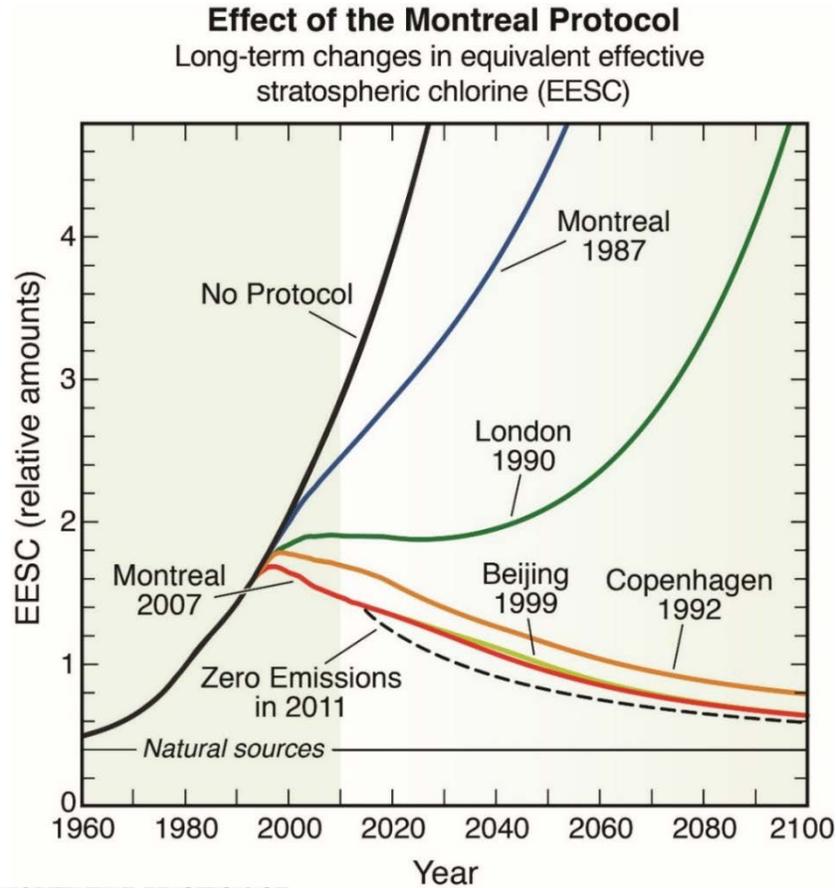
Depletion and Recovery of the Stratospheric Ozone Layer

*Martyn Chipperfield¹, Sandip Dhomse¹, Wuhu Feng¹,
Doug Kinnison², Ryan Hossaini³*

1. University of Leeds, UK. 2. NCAR Boulder, USA 3. Lancaster University, UK

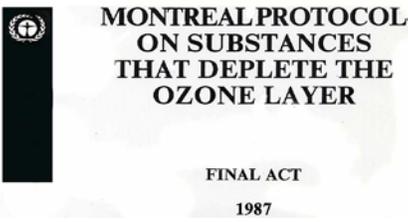
- Ozone recovery?
- World avoided by the Montreal Protocol
- Short-lived anthropogenic chlorine.
- Increasing ozone depletion in the tropics?
- When will 'ozone return' – chemistry-climate simulations

Montreal Protocol (1987)

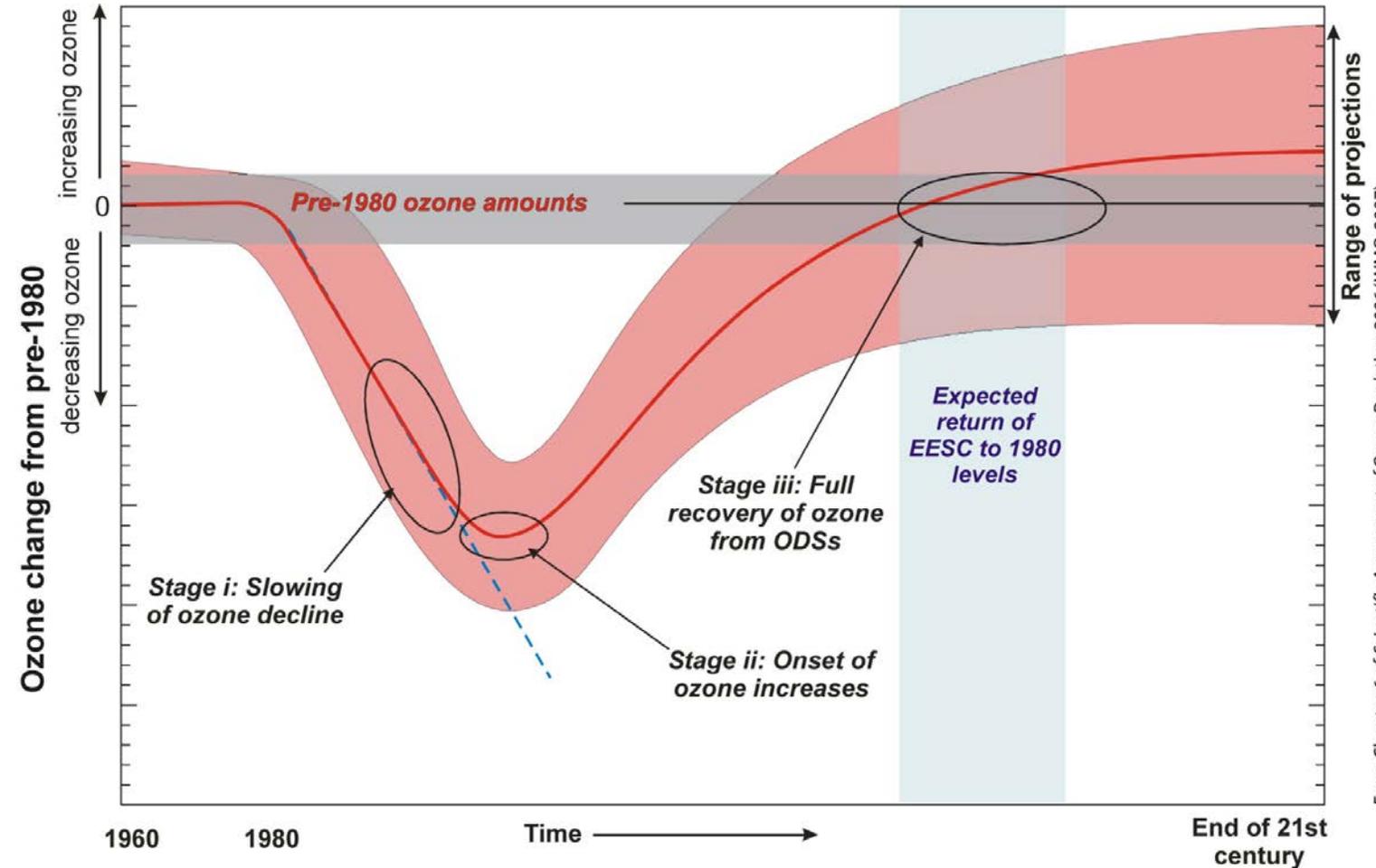


Tropospheric loading of long-lived ozone depleting substances now largely decreasing.

Subsequent decrease in stratospheric chlorine (Cl_y) and bromine (Br_y).



1980 used as reference baseline, but ozone depletion from chlorine and bromine did occur before this time.

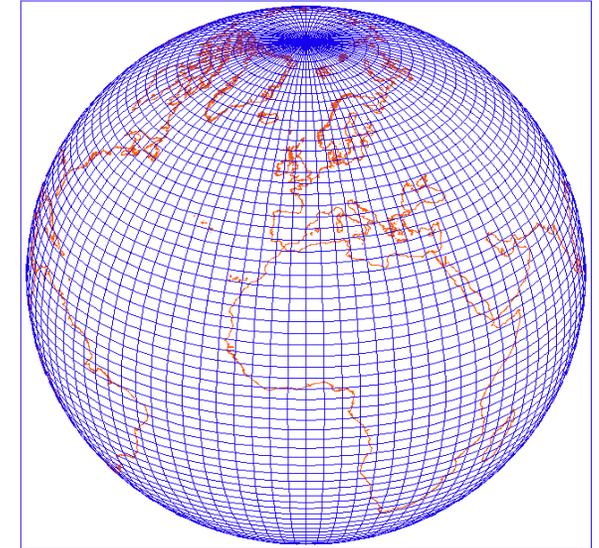


TOMCAT/SLIMCAT 3-D Chemical Transport Model (CTM)

- Used winds and temperatures from meteorological analyses (UK Met Office, ECMWF).
- Chemical species and aerosols transported by winds.
- Chemistry calculated (solves chemical kinetics equations).
- Ideal for simulating past – comparison with specific days.

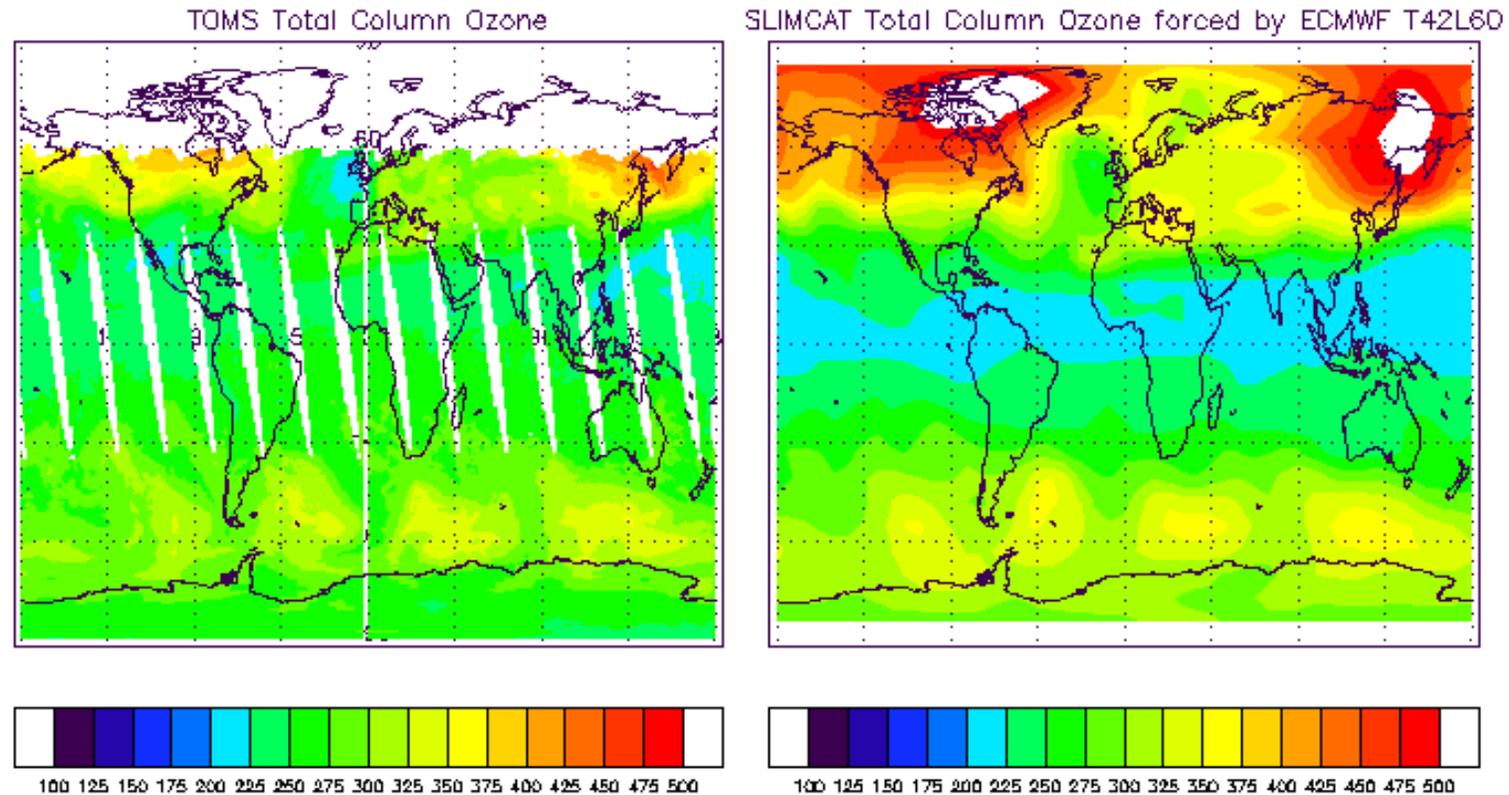
3-D Chemistry-Climate Model (CCM)

- Based on atmospheric general circulation model (GCM).
- Includes chemical species as tracers.
- Include module for calculating chemistry (can be same as CTM).
- Simulates past climatology.
- Needed for future predictions.



TOMS vs SLIMCAT for the Year 2004

2004 1 2



TOMS: Solar backscatter UV observations. Nadir viewing – only column observation.

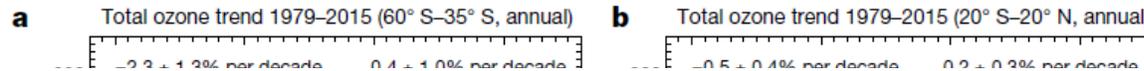


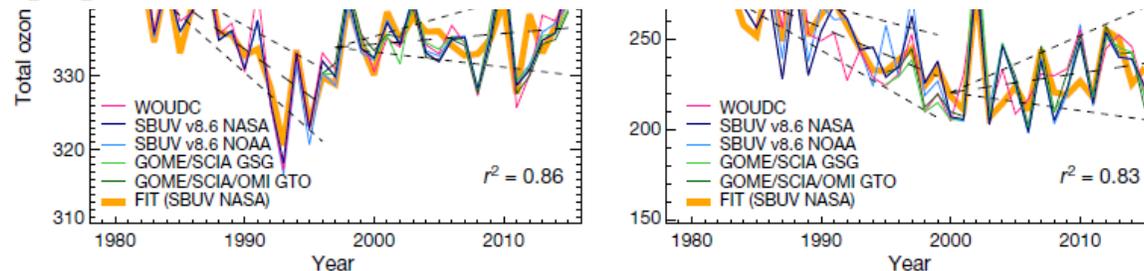
Figure 2 | Time series of observed total (column) ozone. Total ozone is given in Dobson units (DU)

REVIEW

doi:10.1038/nature23681

Detecting recovery of the stratospheric ozone layer

Martyn P. Chipperfield^{1,2}, Slimane Bekki³, Sandip Dhomse¹, Neil R. P. Harris⁴, Birgit Hassler⁵, Ryan Hossaini⁶, Wolfgang Steinbrecht⁷, Rémi Thiéblemont³ & Mark Weber⁸



indicated for the periods before and after the ODS peak (top left and top right, respectively, in each panel), estimated to be in 1996 (middle latitudes and tropics) and 2000 (Antarctic)⁹.

Ozone increasing post 1997, but large uncertainties on the trend.

1. Is ozone recovering?

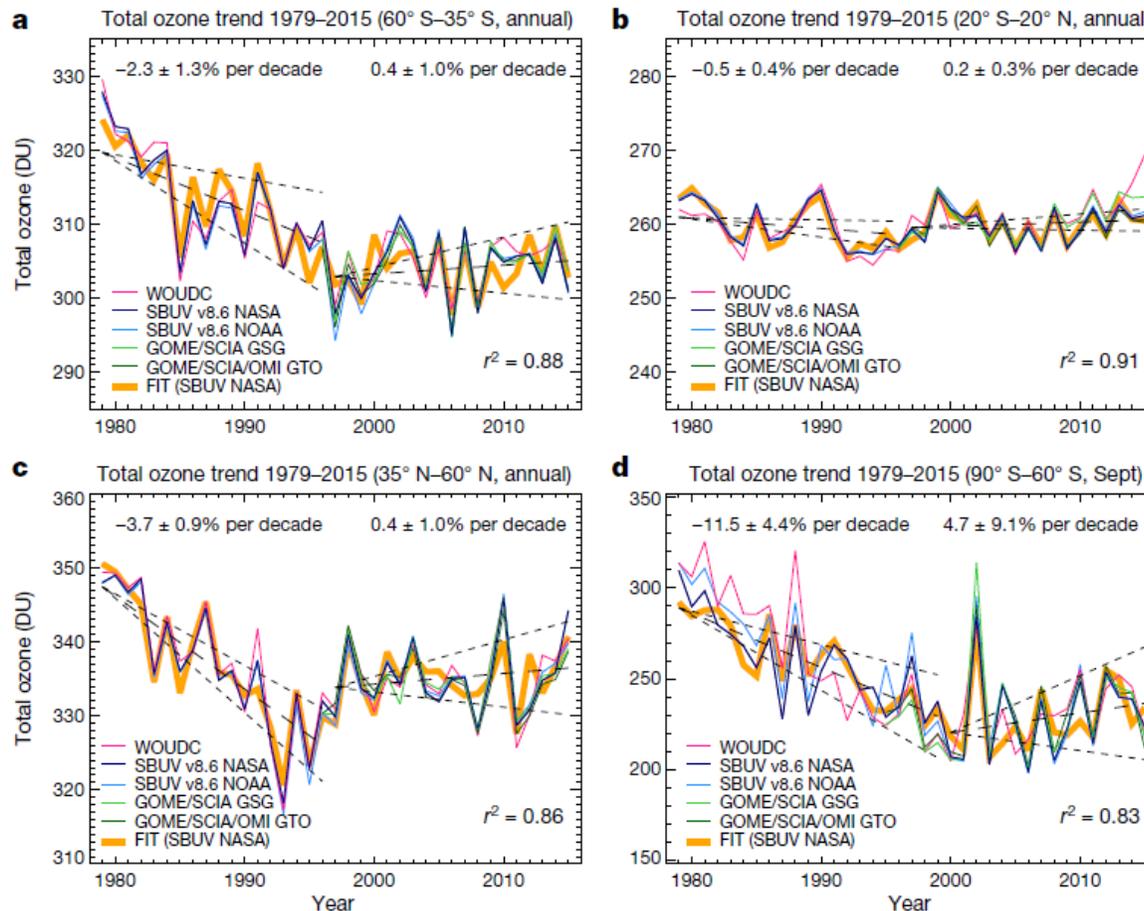


Figure 2 | Time series of observed total (column) ozone. Total ozone is given in Dobson units (DU) for Southern Hemisphere mid-latitudes (a), tropics (b), Northern Hemisphere mid-latitudes (c) and Antarctica in September (d). Shown are time series of the merged SBUV v8.6 data from NOAA⁹² (light blue) and NASA^{21,51} (dark blue), merged GOME/SCIAMACHY/GOME-2 (GSG, light green)³³ and GOME/SCIAMACHY/GOME-2/OMI (GTO, dark green) datasets⁹³ as well as zonal mean data derived from ground-based data collected at the World Ozone and UV Data Center (WOUDC, pink) (updated from ref. 94). Total ozone trends are derived from an MLR applied to the NASA SBUV data and the regression model time series is shown as the orange line. The Supplementary Information gives more details of the MLR approach used here. Linear trends (black long-dashed line) and 2σ uncertainties (black short-dashed lines) as derived from the MLR are indicated for the periods before and after the ODS peak (top left and top right, respectively, in each panel), estimated to be in 1996 (middle latitudes and tropics) and 2000 (Antarctic)⁹.

Ozone increasing post 1997, but large uncertainties on the trend.

Model setup:

- Detailed stratospheric chemistry scheme. Specified aerosol SAD (3λ).
- Forced by ECMWF ERA-Interim winds and temperatures (no feedback).
- 1955 – 2015 (repeating 1980 meteorology before 1980)
- $2.8^\circ \times 2.8^\circ$. 32 levels: surface – 60 km.

Six simulations:

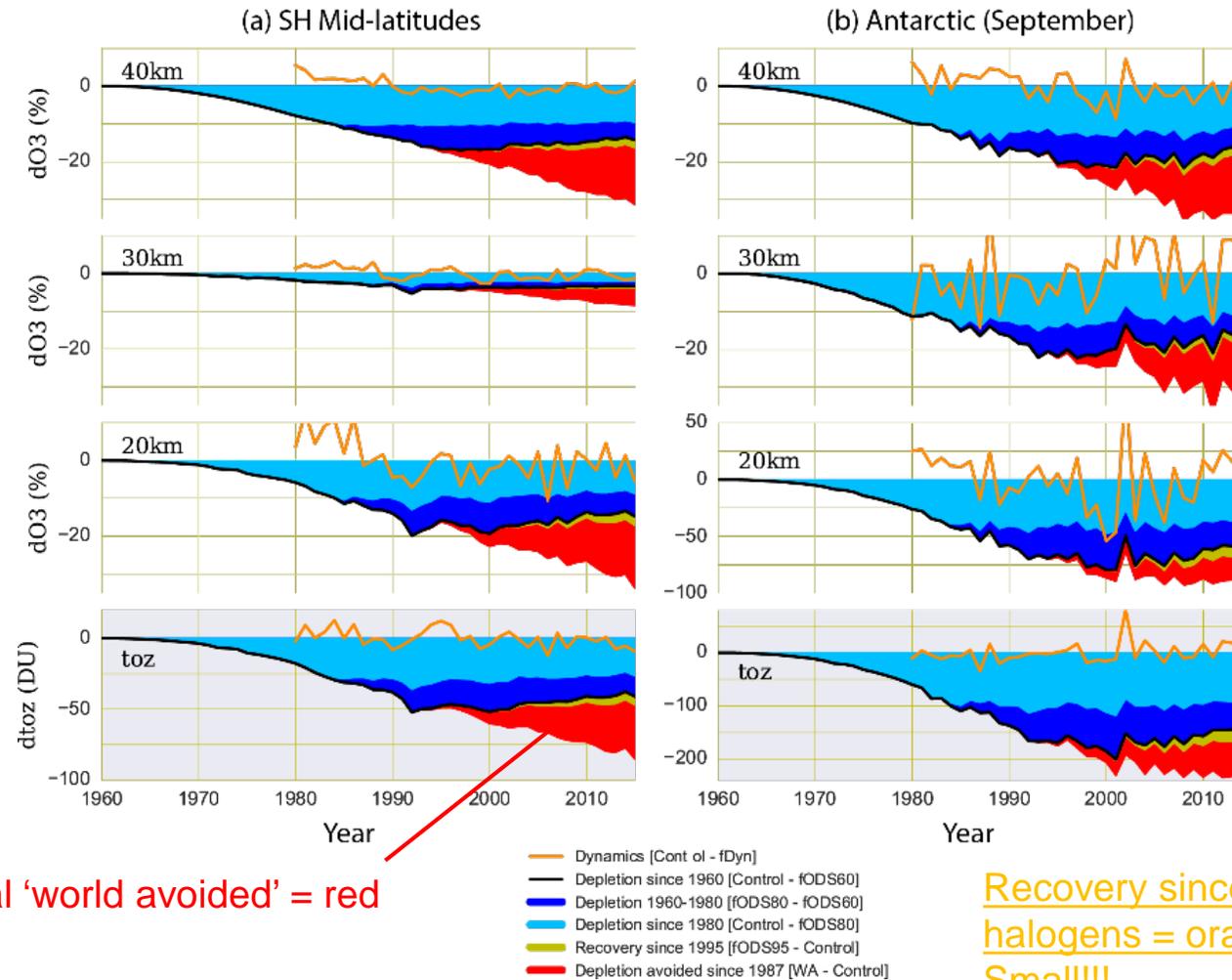
- Control. Forced by observed surface mixing ratios of long-lived ODSs (CFCs, HCFCs, solvents, CH_3Cl ...)
- **fODS 1960**. ODSs fixed at 1960 tropospheric vmr
- **fODS 1980**.
- **fODS 1995**. (Peak tropospheric halogen: chlorine + α bromine).
- **World avoided**: 3%/year growth in ODS emissions after 1987.
- **Repeating 1980** meteorology.

Quantifying ozone loss and recovery



3D model

Contributions to modelled ozone depletion since 1960



Actual depletion = blue + dark blue

Theoretical 'world avoided' = red

Recovery since peak halogens = orange
Small!!!!

2002: Small ozone hole due to split vortex. 2015: Large ozone hole due to volcanic aerosol

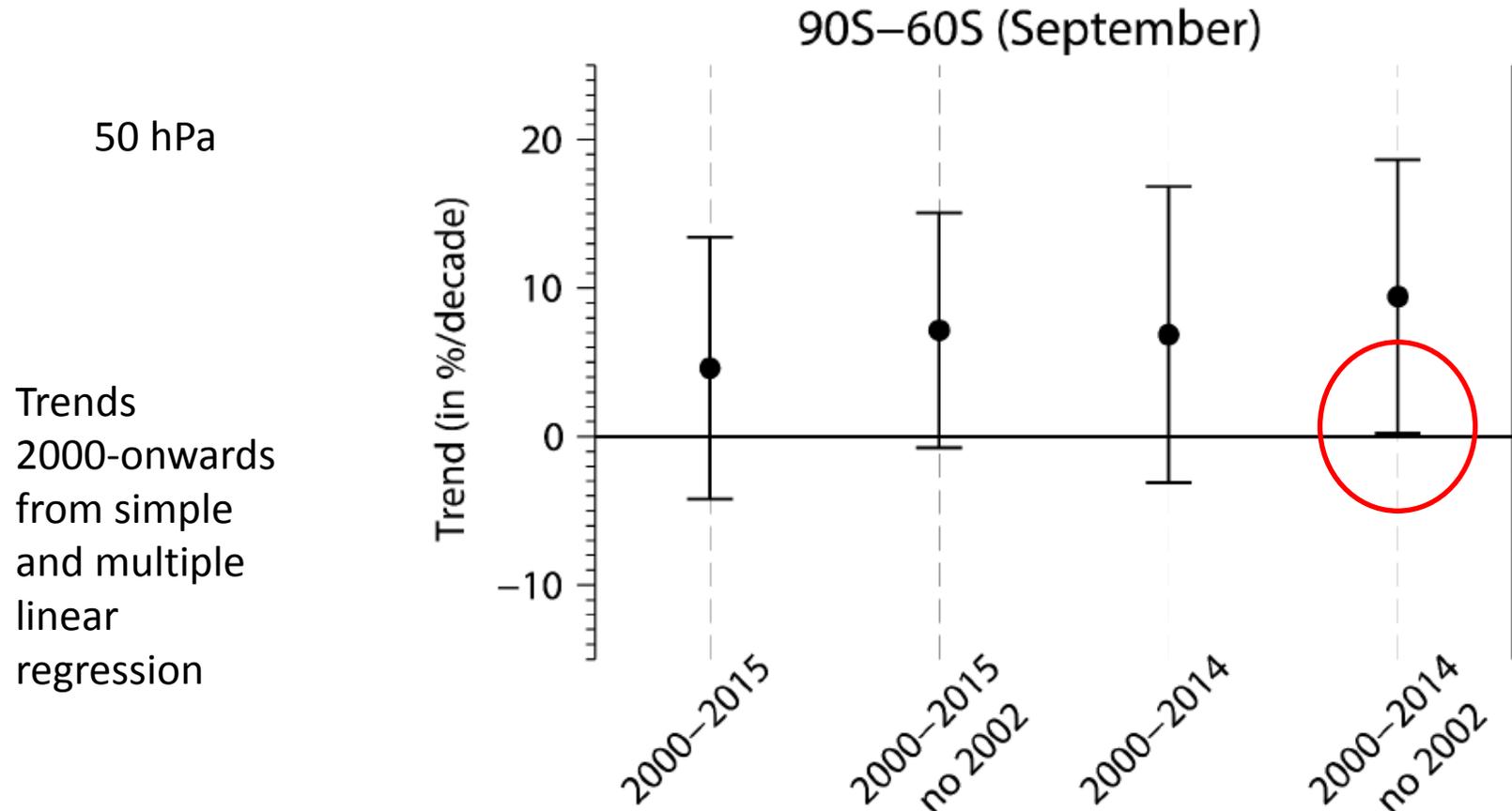
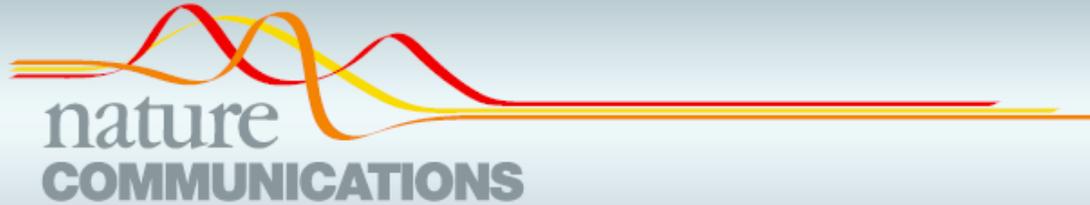


Figure S1. Modelled Antarctic total ozone trends (%/decade) from multiple linear regression (MLR) for the control (varying ODS and dynamics) simulation. The different trends correspond to different periods (2000–2015, 2000–2014) with and without the year 2002. The error bar indicates the 2σ uncertainty. Note that Solomon et al.¹⁴ did not include the year 2002 in their 2000–2014 trend analysis. The 2000–2015 trend is also shown in the bottom left panel of **Figure 5**.

Remi Thieblemont
and Slimane Bekki



ARTICLE

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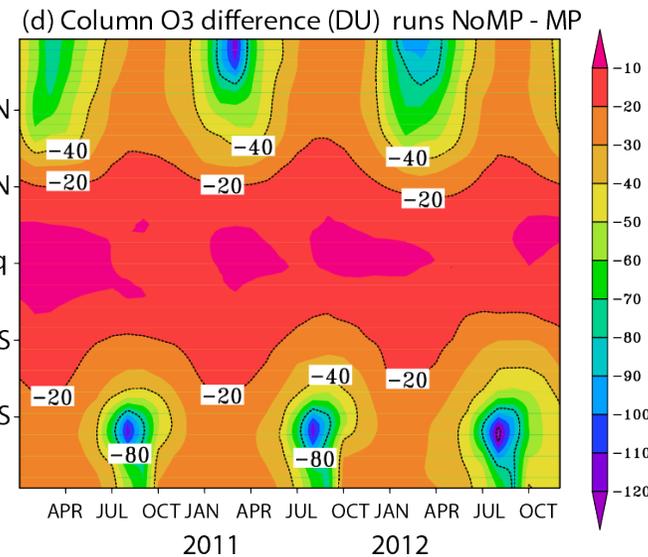
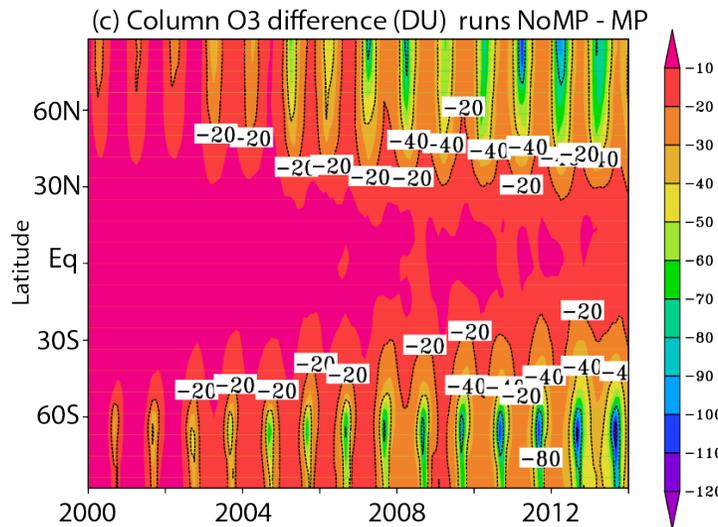
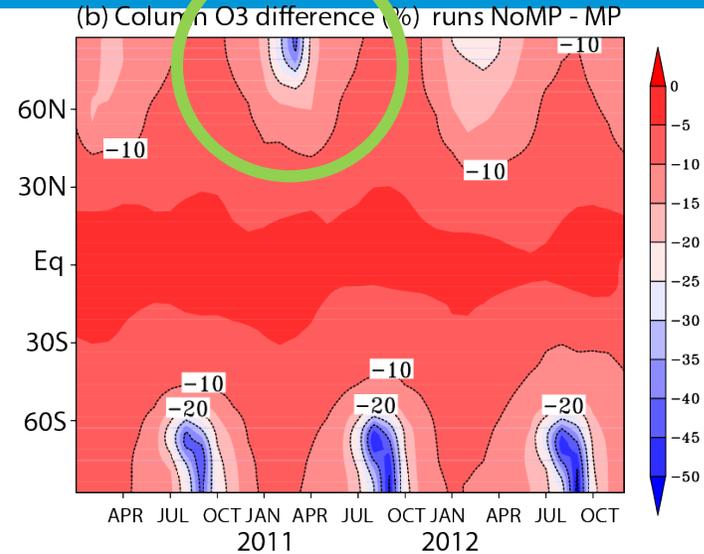
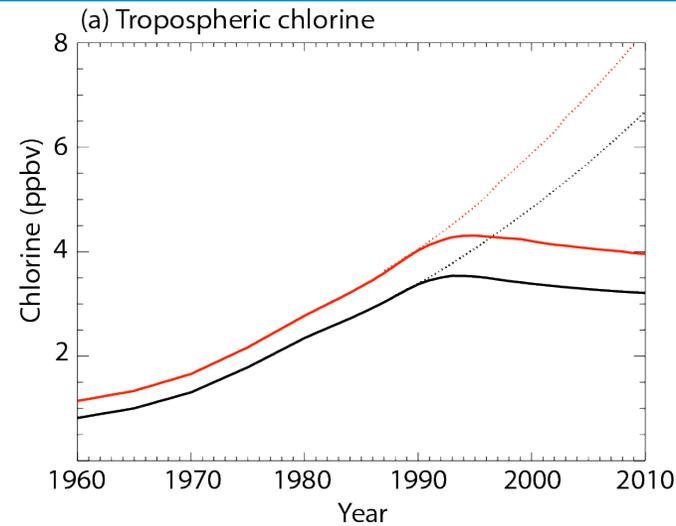
DOI: [10.1038/ncomms8233](https://doi.org/10.1038/ncomms8233)

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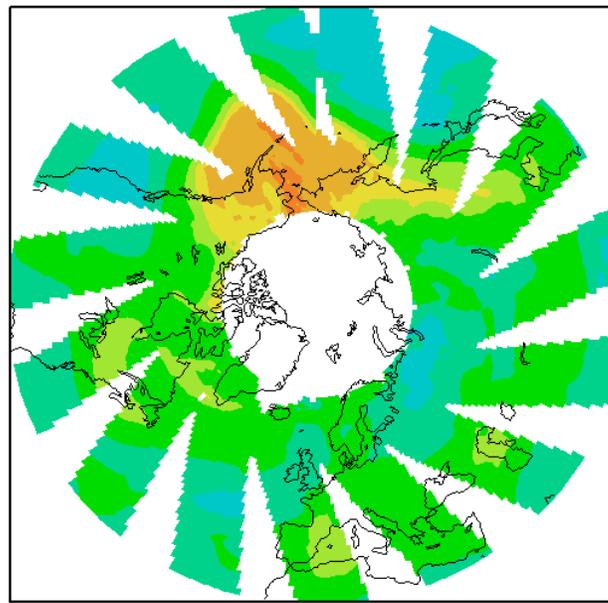
Quantifying the ozone and ultraviolet benefits already achieved by the Montreal Protocol

M.P. Chipperfield^{1,2}, S.S. Dhomse^{1,2}, W. Feng^{1,3}, R.L. McKenzie⁴, G.J.M. Velders⁵ & J.A. Pyle^{3,6}

Column ozone difference with/without MP



OMI TOZ:01NOV2010

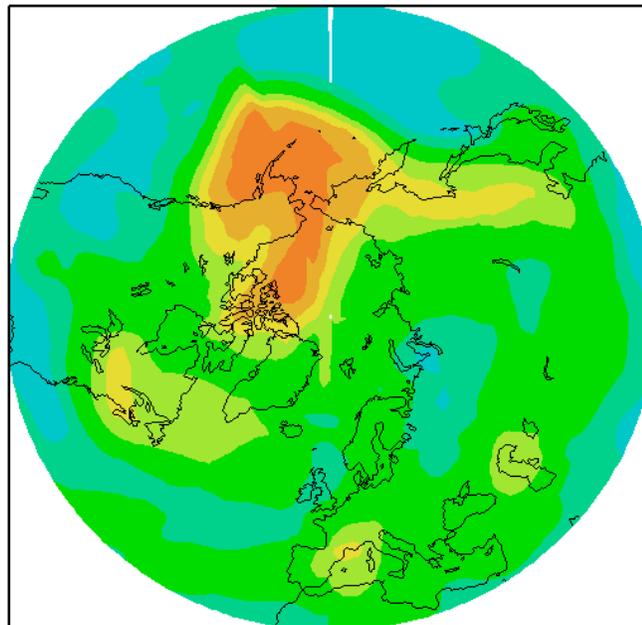


Satellite observations

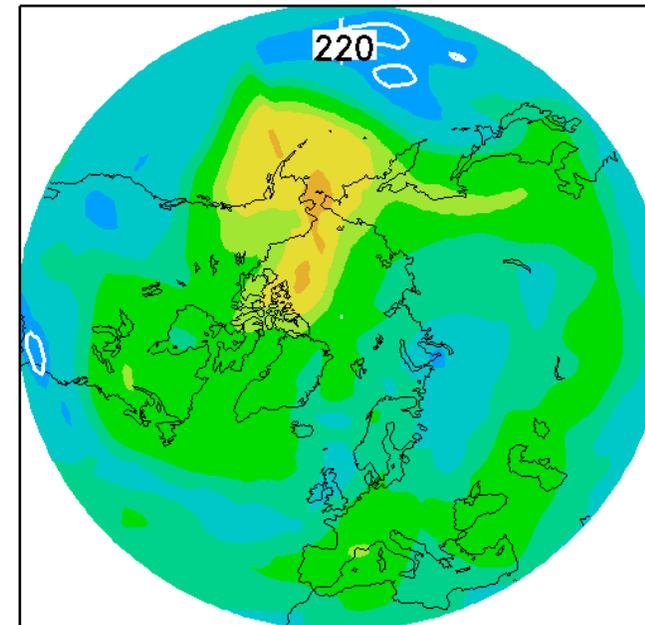
With Montreal Protocol

No Montreal Protocol

MP TOZ:01NOV2010



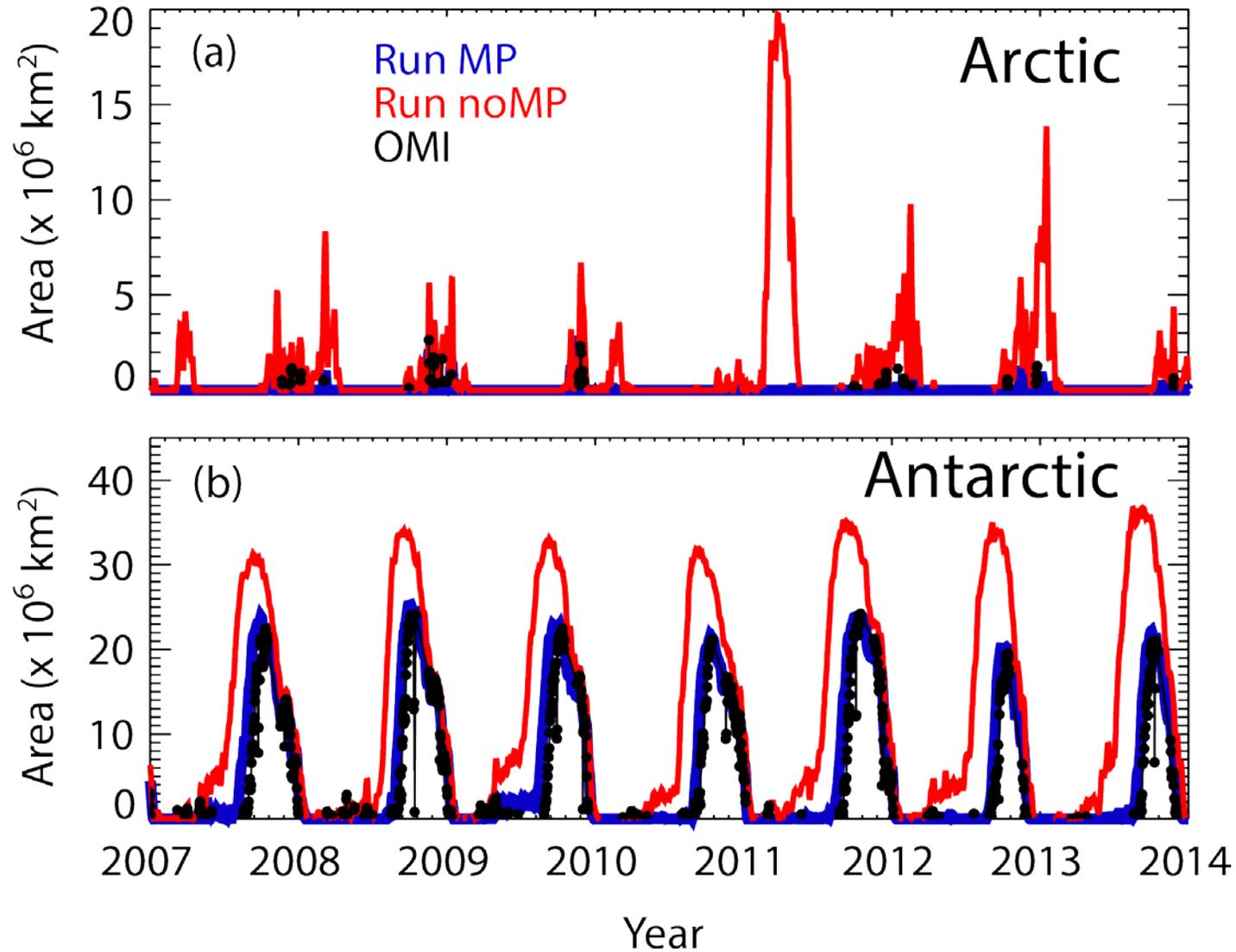
NoMP TOZ:01NOV2010

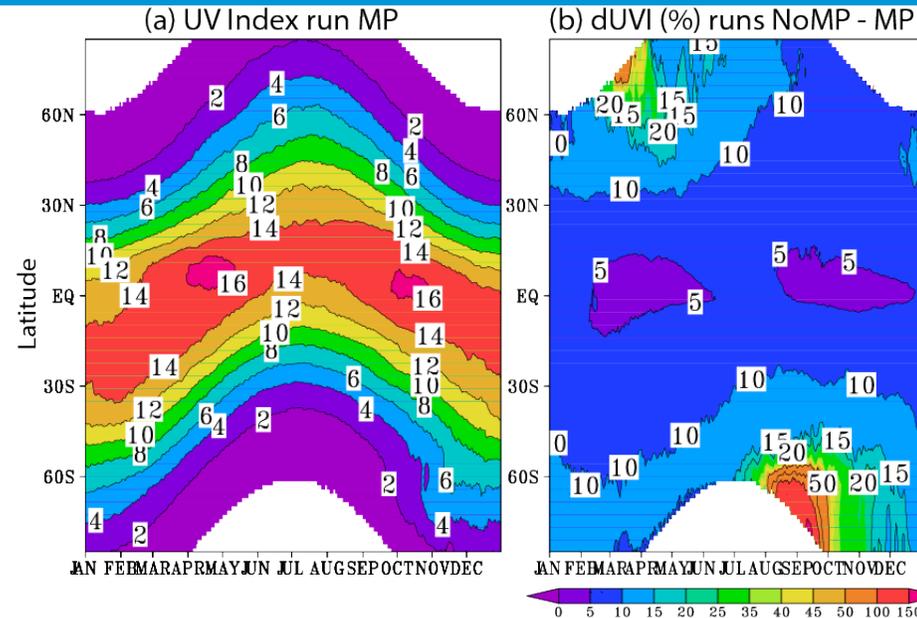


Area of 'Arctic' and Antarctic ozone holes

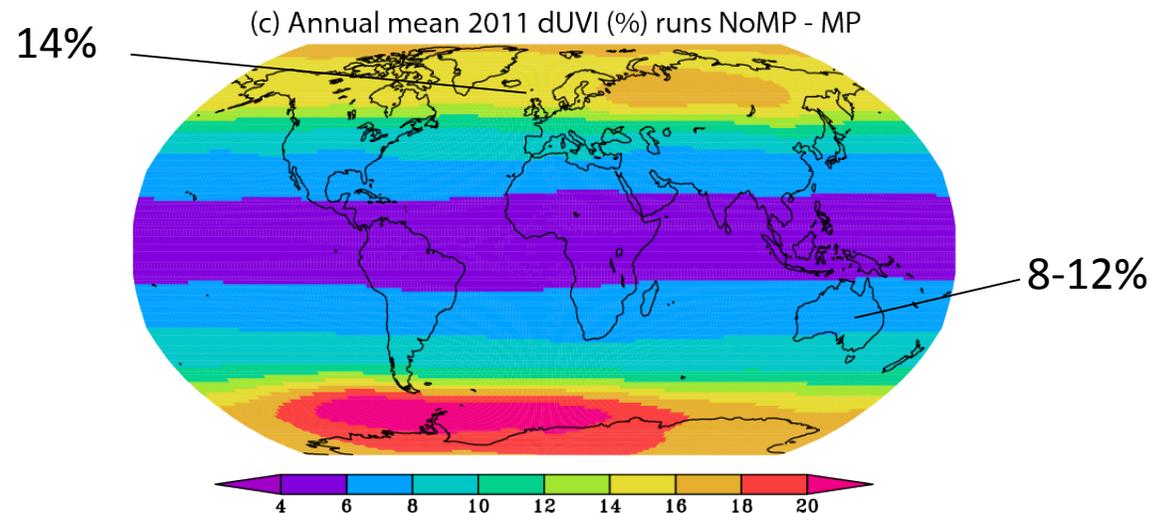


Area within
220 DU
contour





5% increase in sunburning UV could cause increases of: 15% (squamous) and 8% (basal) cell carcinomas (Longstreth et al. 1998).





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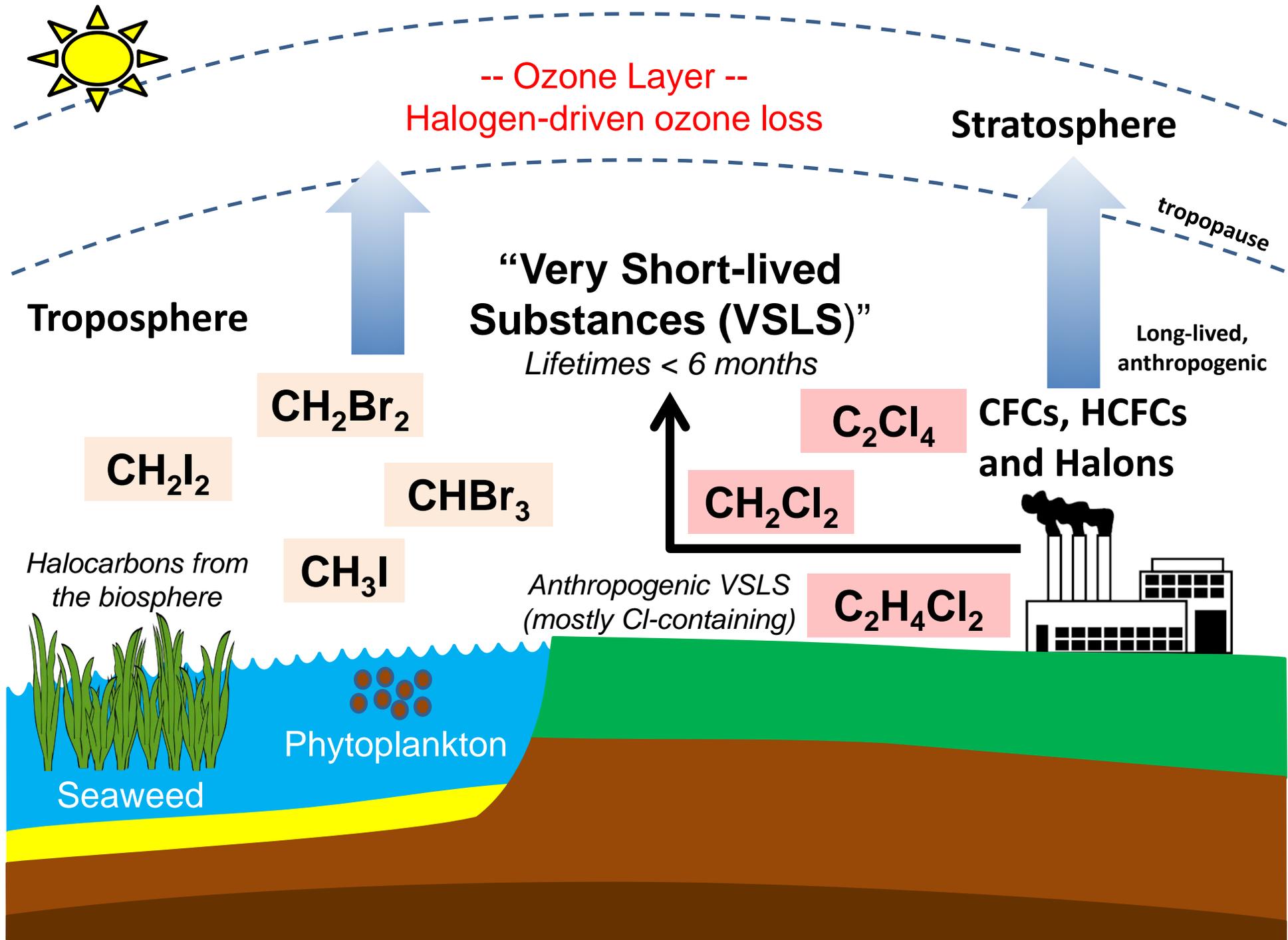
Received 16 Oct 2016 | Accepted 16 May 2017 | Published 27 Jun 2017

DOI: [10.1038/ncomms15962](https://doi.org/10.1038/ncomms15962)

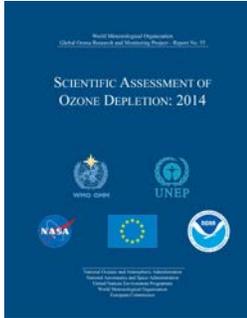
OPEN

The increasing threat to stratospheric ozone from dichloromethane

Ryan Hossaini¹, Martyn P. Chipperfield^{2,3}, Stephen A. Montzka⁴, Amber A. Leeson¹, Sandip S. Dhomse^{2,3}
& John A. Pyle^{5,6}



2014 Report



$$\text{Br}_y^{\text{VSLS}} = 2 - 8 \text{ ppt}$$

$$\text{Cl}_y^{\text{VSLS}} = 95 (50-145)$$

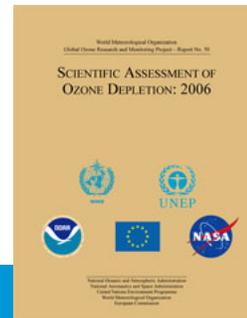
2010 Report



$$\text{Br}_y^{\text{VSLS}} = 1 - 8 \text{ ppt}$$

$$\text{Cl}_y^{\text{VSLS}} = 80 (40-130)$$

2006 Report



$$\text{Br}_y^{\text{VSLS}} = 3 - 8 \text{ ppt}$$

Best estimates from recent WMO/UNEP Stratospheric Ozone Assessments

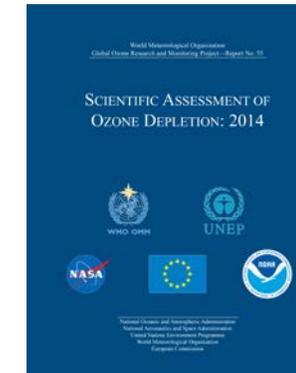
- VSLS account for ~25% of strat. Br_y (i.e. 5 ppt out of a total of ~20 ppt in 2011)
- VSLS make a much smaller % contribution of total strat. Cl_y (i.e. 95 ppt out of several thousand ppt)

Stratospheric Cl from VSLS



Observed values in the tropical upper troposphere

VSLs	Source	VMR (ppt)	Range (ppt)
CH ₂ Cl ₂	Anthropogenic	23.2	17.5 – 27.8
CHCl ₃	Anthrop. + natural	5.7	3.5 – 7.9
C ₂ H ₄ Cl ₂	Anthropogenic	2.7	1.6 – 4.9
C ₂ HCl ₃	Anthropogenic	0.03	0.0 – 0.17
C ₂ Cl ₄	Anthropogenic	0.7	0.3 – 1.3

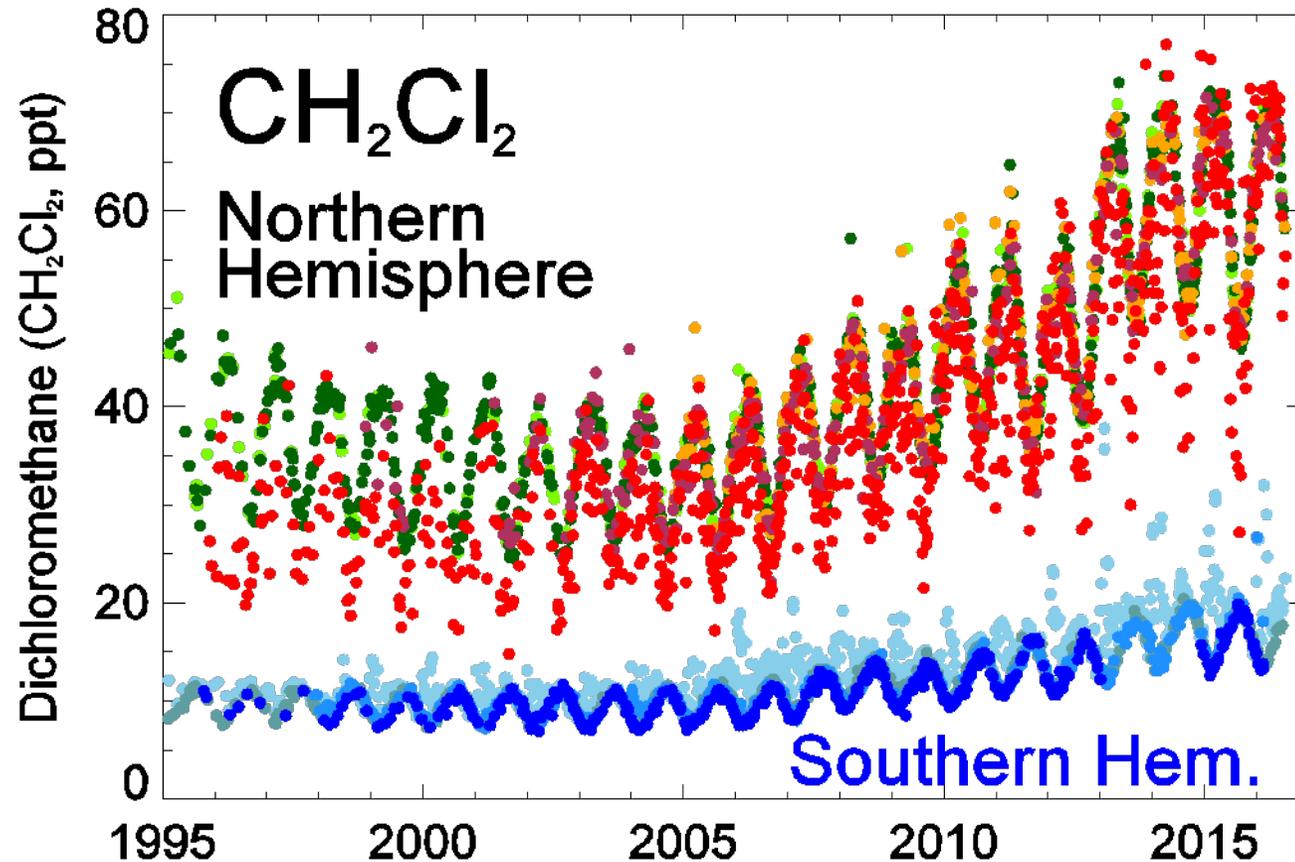


Σ Cl in Source Gases = 72 (50 – 95) ppt Cl

Σ Cl in Product Gases = 25 (0 – 50) ppt Cl

Total Cl = 95 (50 – 145) ppt Cl

Growth in CH₂Cl₂ (dichloromethane)



Surface CH₂Cl₂
>doubled in the
last decade

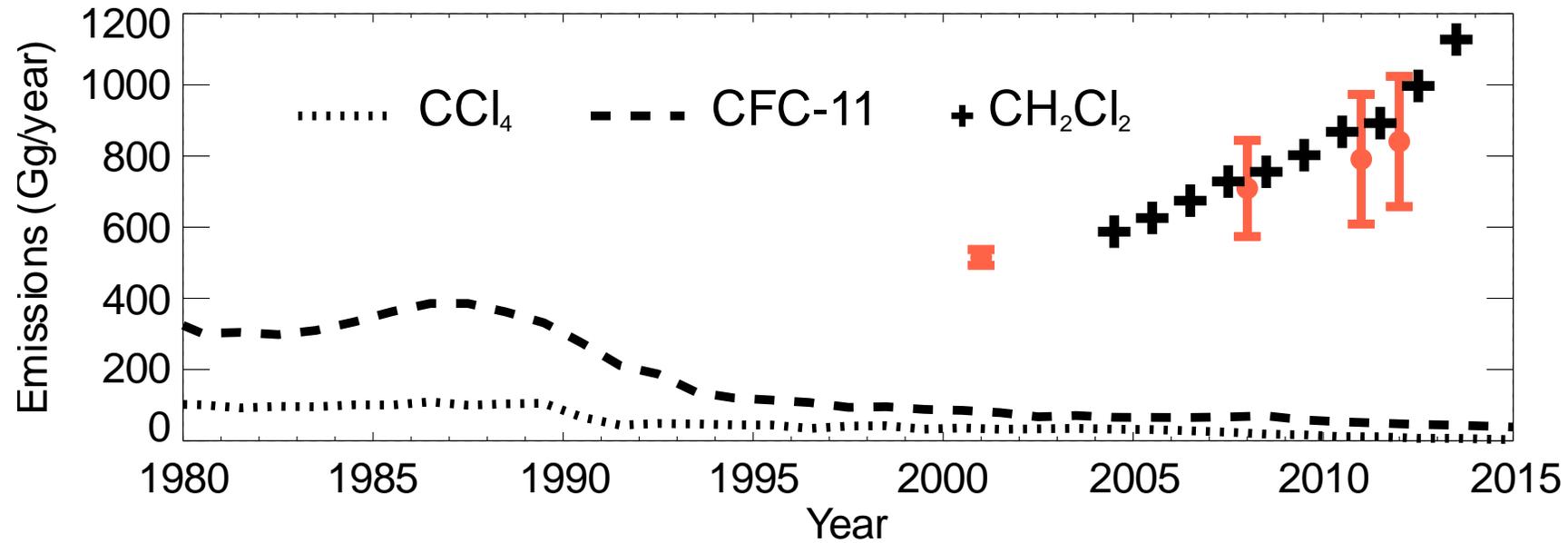
Mean growth
rate in NH of
~8% per year

Not controlled
by Montreal
Protocol



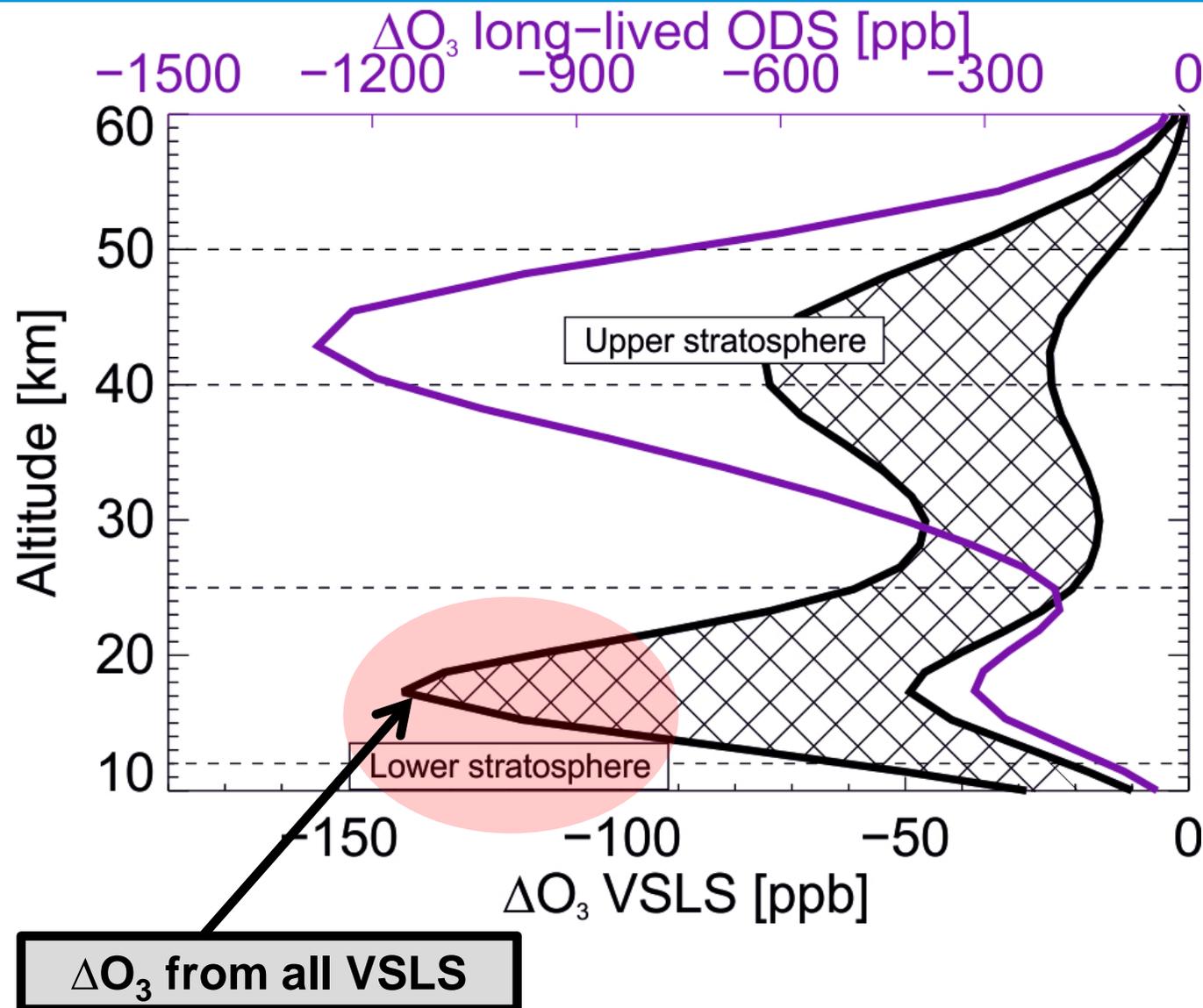
Applications include: Metal cleaning, paint remover, use as a chemical feedstock (including for certain HFCs)

CH₂Cl₂ global emissions



- CH₂Cl₂ emissions (+) large relative to long-lived ODSs in 1980s..
- .. though CH₂Cl₂ has a much much shorter lifetime

Altitude-resolved ΔO_3 due to VSLS



ΔO_3 from long-lived gases (CFCs etc.)

SLIMCAT 3-D CTM with/without VSLS (Br + Cl + I)

Grated area is uncertainty due to strat. VSLS loading.

Per molecule, O_3 perturbations in UTLS cause greatest radiative effect

Altitude-resolved ΔO_3 due to VSLs

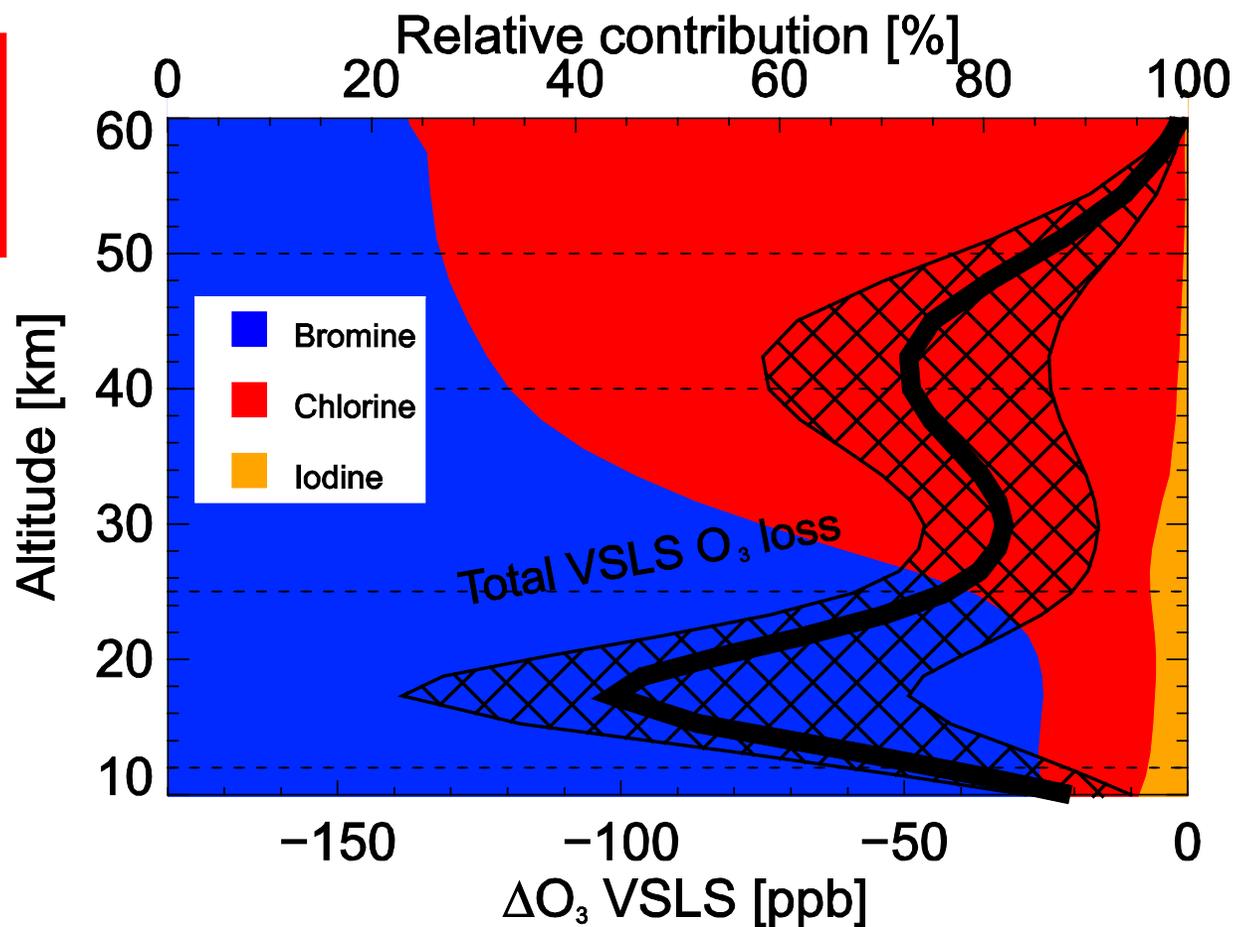


Natural bromine VSLs are the most important for stratospheric O_3

Relative contribution to LS O_3 loss from VSLs

- Bromine (86%)
- Chlorine (11%)**
- Iodine (3%)

Chlorine-containing VSLs are of mostly anthropogenic origin



VSLs Tracers

Natural

CHBr₃
CH₂Br₂
CHBr₂Cl
CH₂BrCl
CHBrCl₂
CH₃I

Anthrop.

CHCl₃
CH₂Cl₂
C₂Cl₄
C₂H₄Cl₂
C₂HCl₃

**TOMCAT/SLIMCAT
3-D CTM**

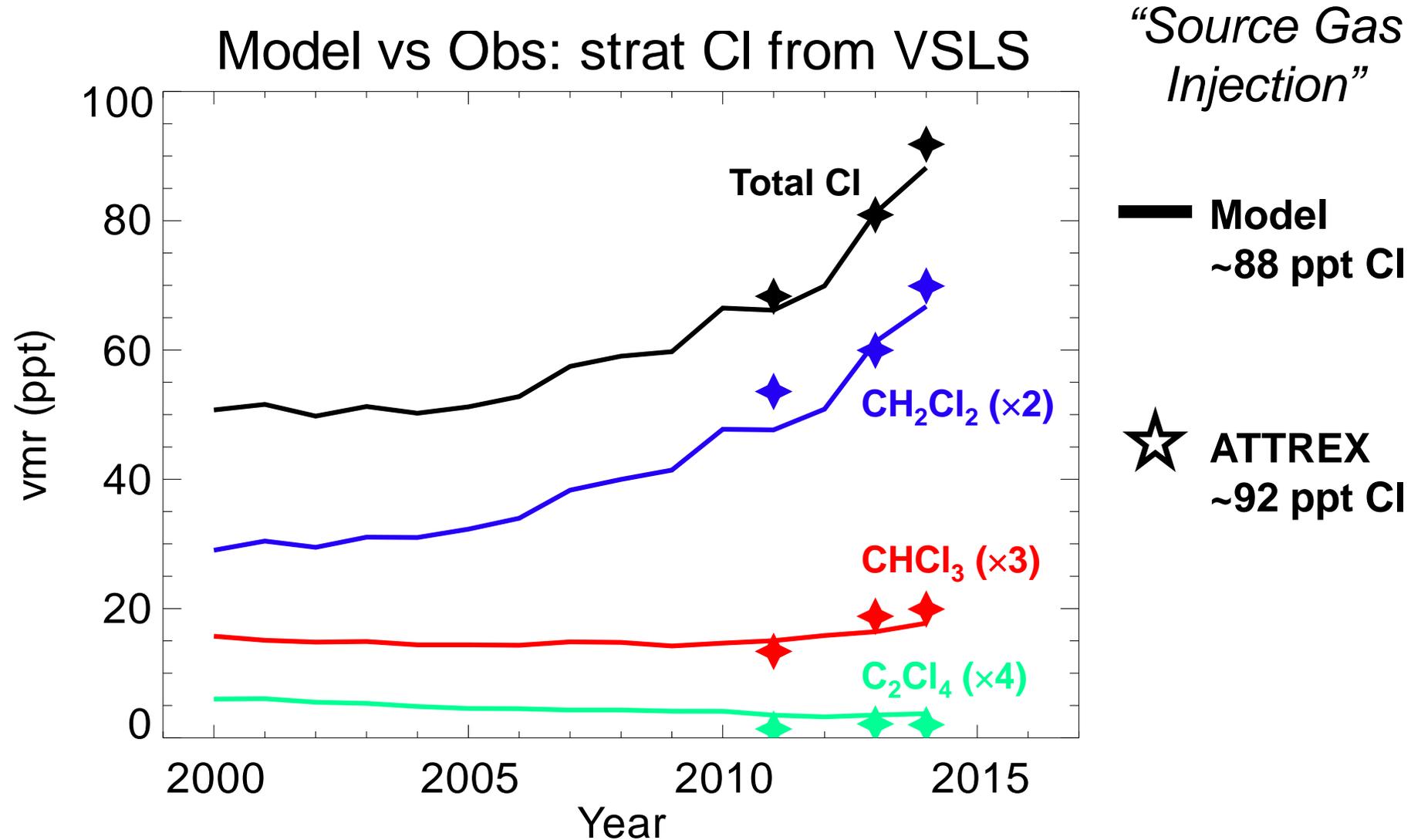
Quantify stratospheric
Cl from VSLs
(2000 - 2014)

*Simulations 1980-2100
Repeating meteorology in future*

With and without CH₂Cl₂ growth

**Isolate the impact of
CH₂Cl₂ growth on
ozone recovery**

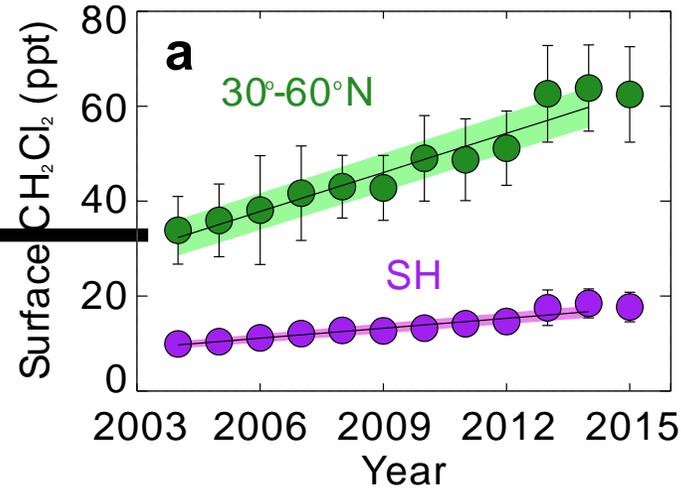
Time-dependent surface
boundary condition based on
NOAA/AGAGE long term data
(5 latitude bands)



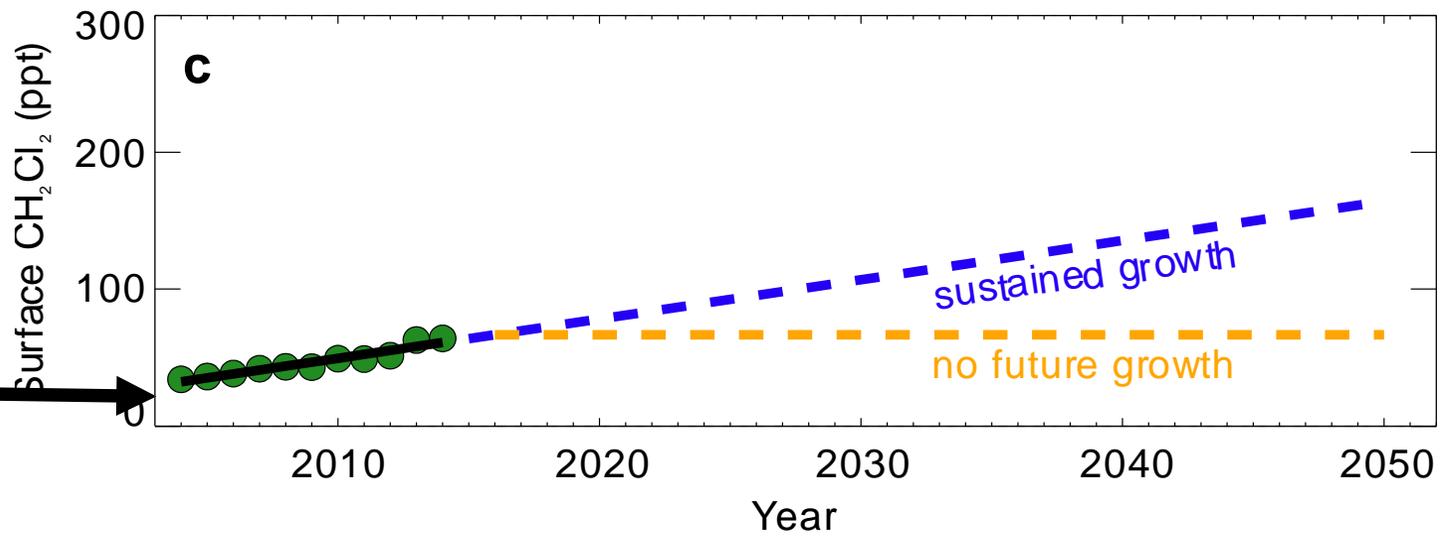
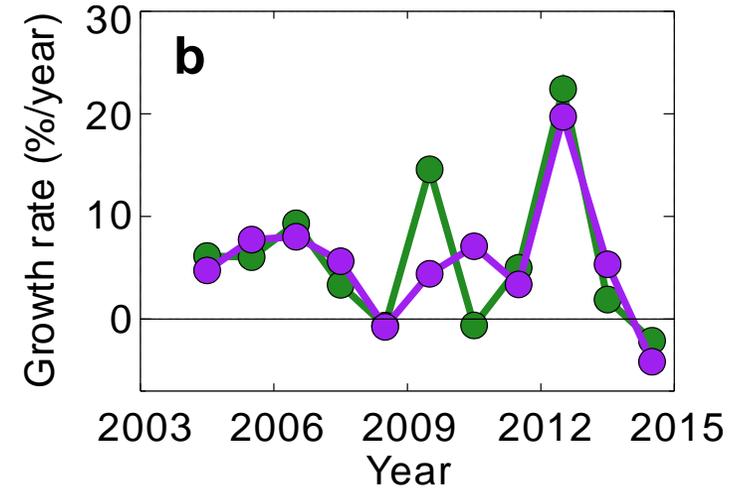
CH₂Cl₂ growth scenarios



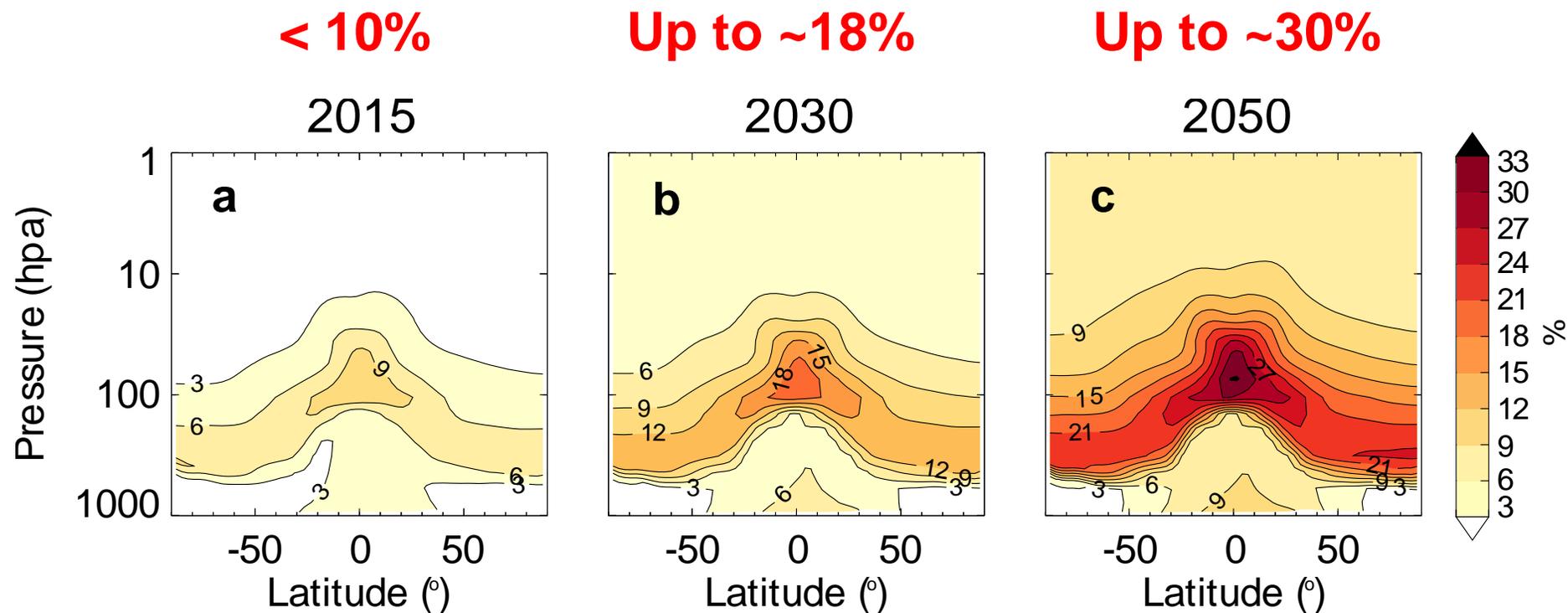
Observed surface CH₂Cl₂ mole fraction (ppt)



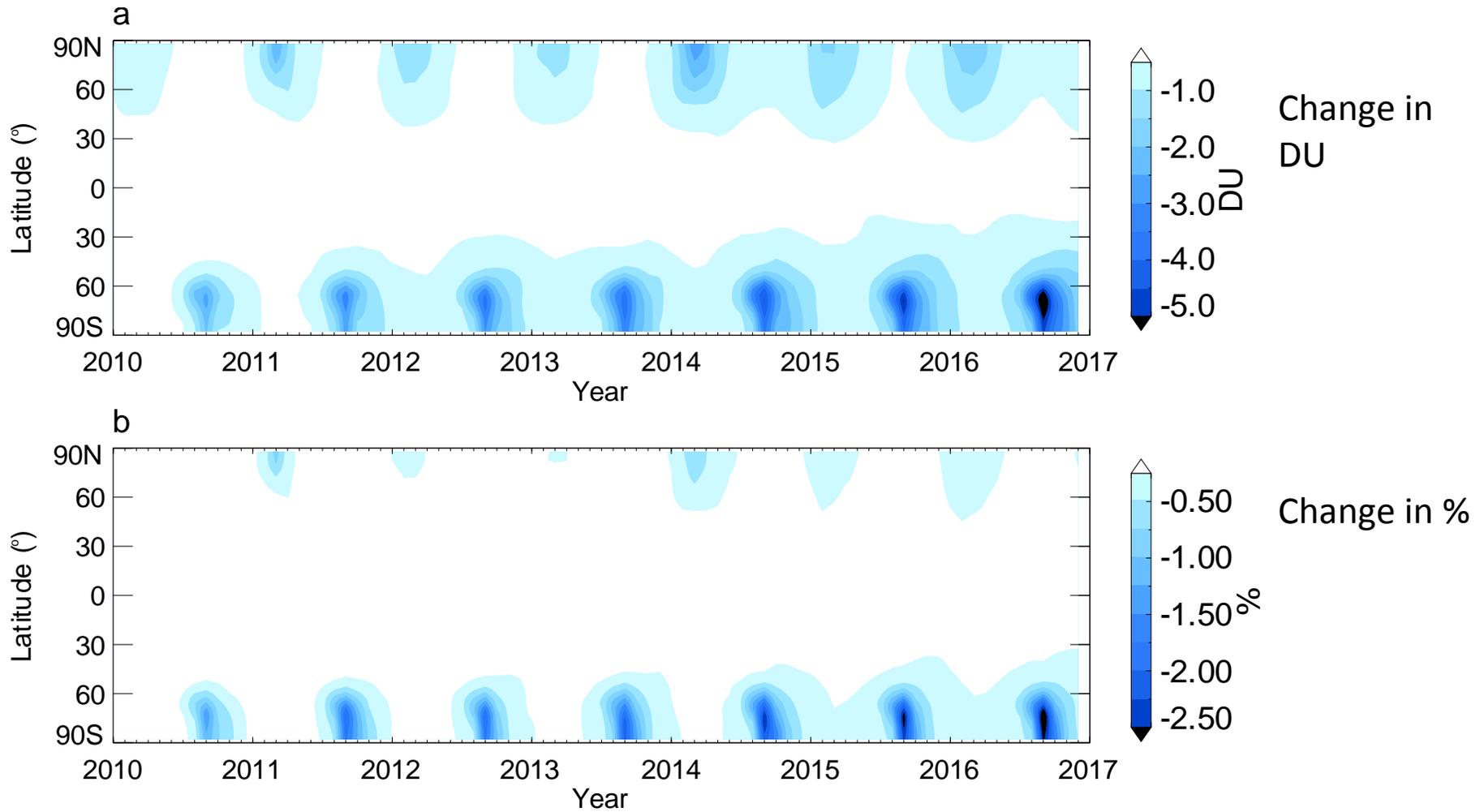
Observed surface CH₂Cl₂ growth rates (% per year)



Contribution of CH₂Cl₂ to strat. Cly

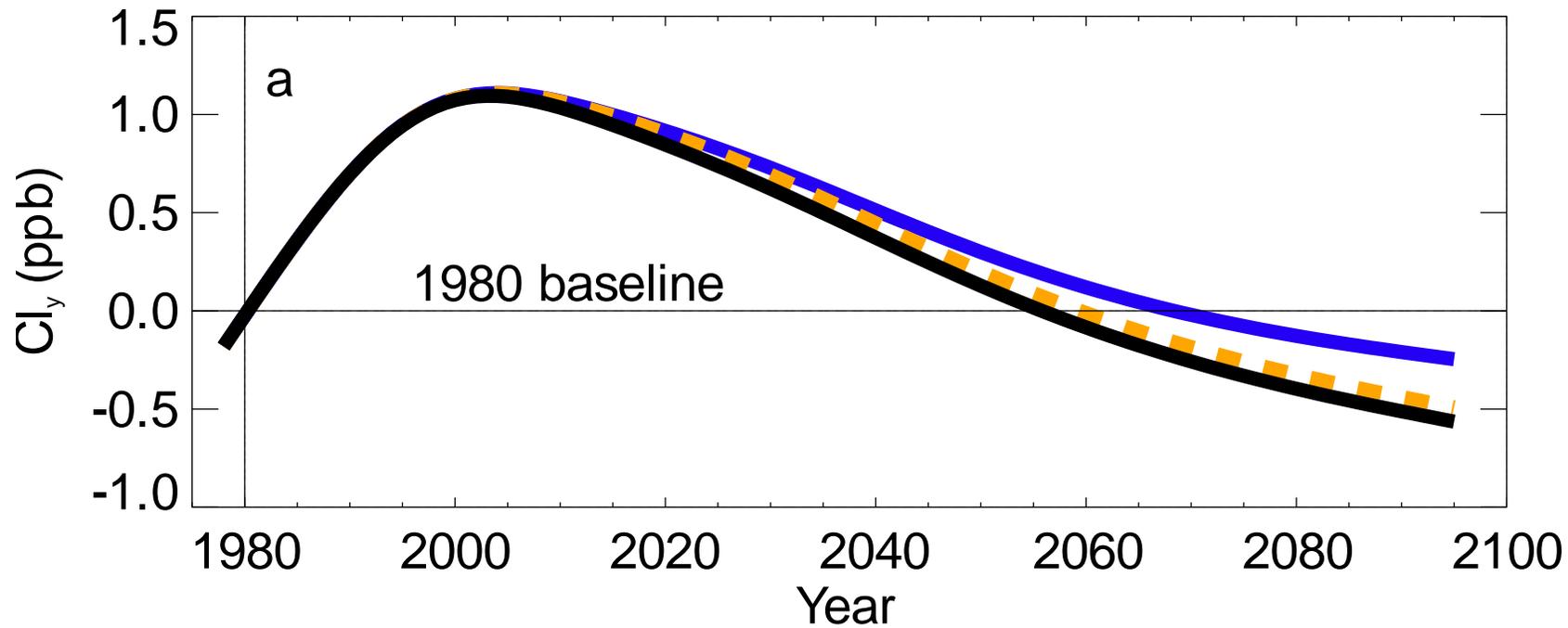


Impact of CH₂Cl₂ on O₃ in recent past



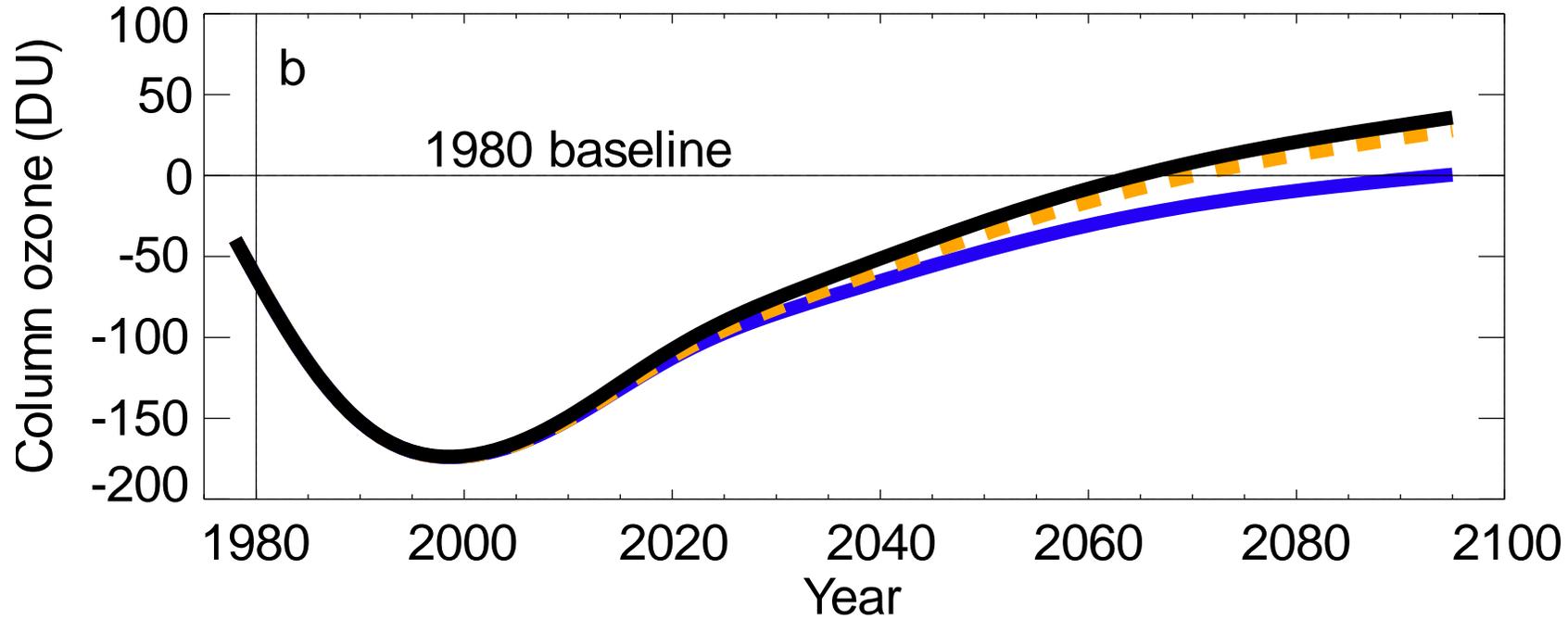
The impact of CH₂Cl₂ on ozone has already significantly increased over the last decade

Impact of CH₂Cl₂ growth: Antarctic Cl_y



Experiment	Return date
No CH ₂ Cl ₂	2056
w/ CH ₂ Cl ₂ #1	2060 (no future growth)
w/ CH ₂ Cl ₂ #2	2069 (sustained future growth)

Impact of CH₂Cl₂ growth: Antarctic O₃



Experiment	Return date
No CH ₂ Cl ₂	2065
w/ CH ₂ Cl ₂ #1	2071 (no future growth)
w/ CH ₂ Cl ₂ #2	2095 (sustained future growth)

4. Is tropical ozone decreasing?



6

W. T. Ball et al.: Continuous stratospheric ozone decline

ACP
2018

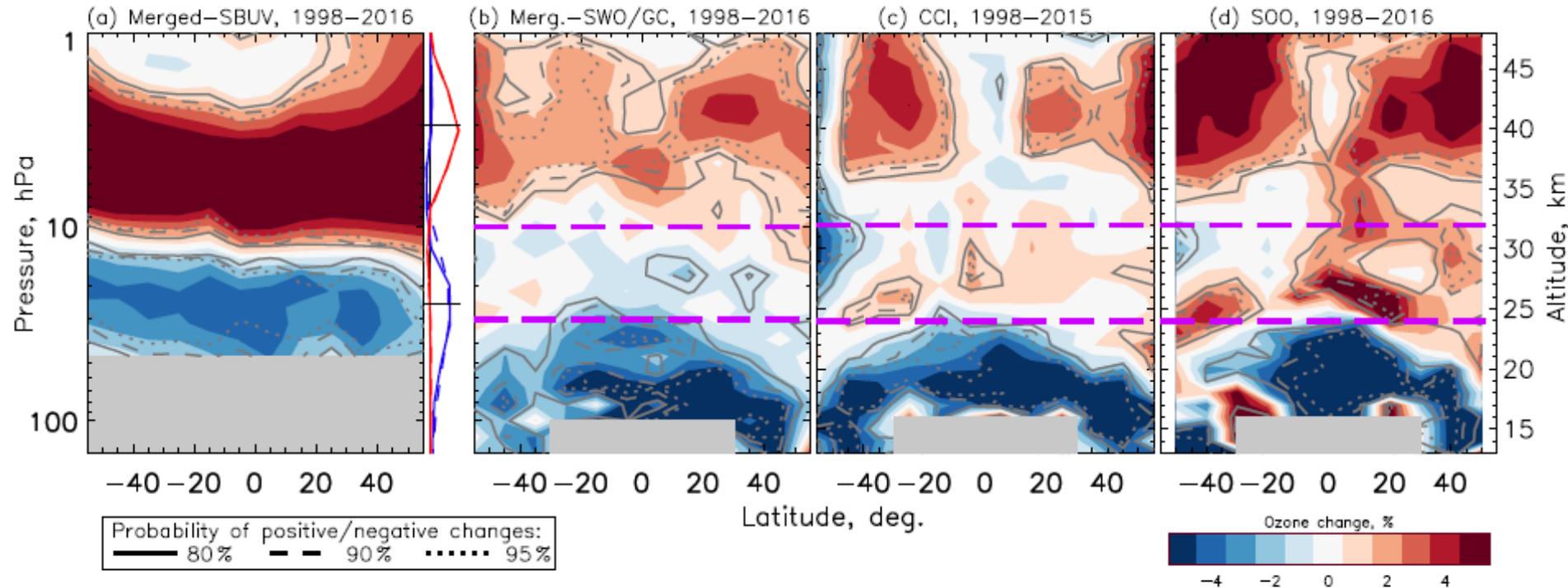


Figure 1. Zonally averaged change in ozone between 1998 and 2016. From (a–d) the Merged-SBUV, Merged-SWOOSH/GOZCARDS, SAGE-II/CCI/OMPS (CCI), and SAGE-II/OSIRIS/OMPS (SOO) composites. Red represents increases, blue decreases (%; see right legend). Contours represent probability levels of positive or negative changes (see left legend). Grey shaded regions represent unavailable data. Pink dashed lines delimit regions integrated into partial ozone columns in Fig. 2 (and Figs. S3, S4, S6, S9, and S10). To the right of Merged-SBUV are the instrument observing profiles centred at 3 hPa (red, upper) and 25 hPa (blue) at northern mid-latitudes (dashed) and in the tropics (solid) from Kramarova et al. (2013). SAGE-II/CCI/OMPS changes are for 1998–2015.

Is tropical ozone decreasing?



O₃ decreasing?
Models fail to capture this.
Due to short-lived species?
Montreal Protocol not working?

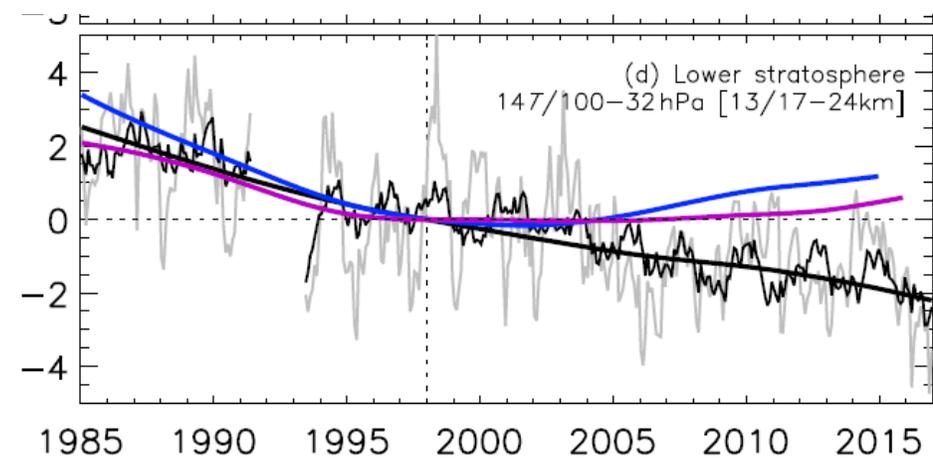
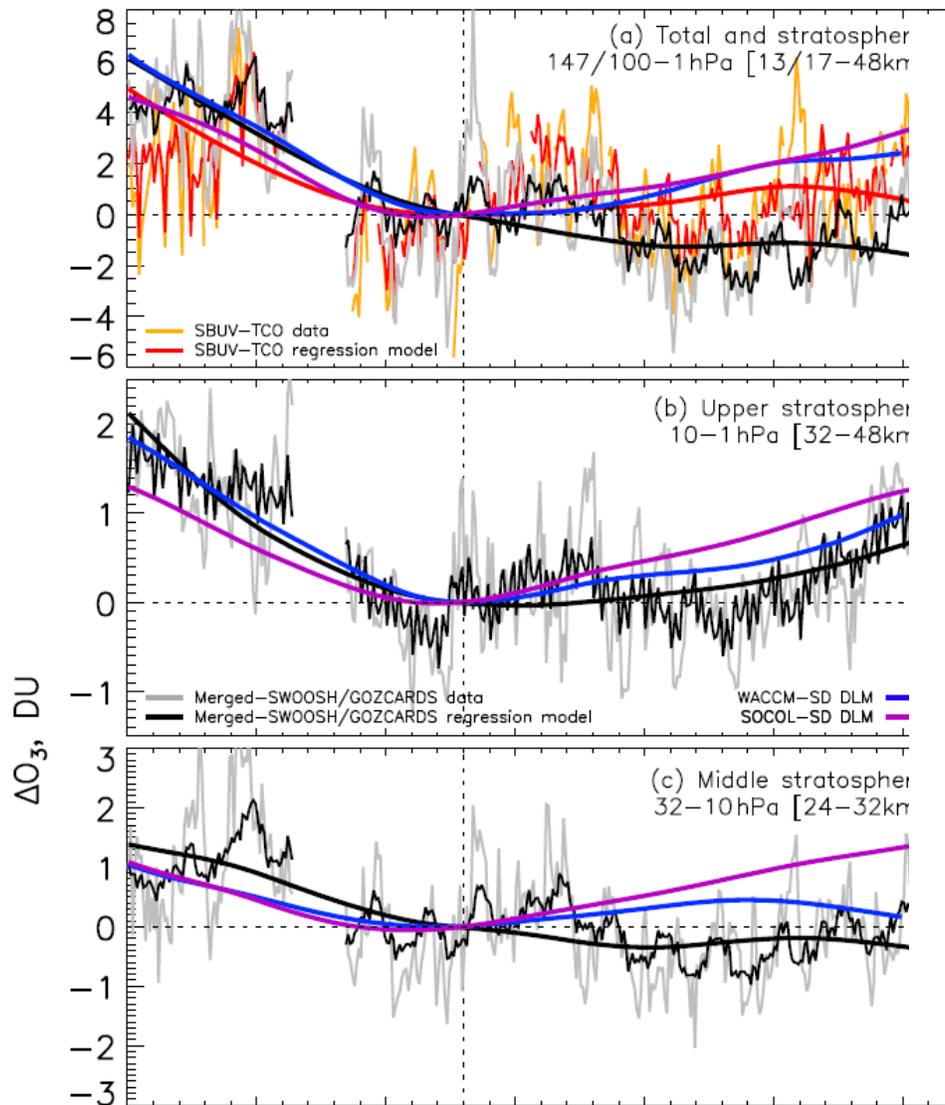
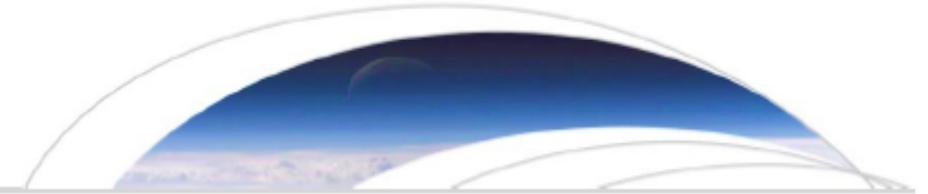


Figure 3. Total and partial column ozone anomalies integrated over 60° S–60° N between 1985 and 2016. Deseasonalised and regression model time series are given for the Merged-SWOOSH/GOZCARDS composite (grey and black, respectively) for (a) the whole stratospheric column and (b) upper, (c) middle, and (d) lower stratospheric partial column ozone. The DLM non-linear trend is the smoothly varying thick black line. In (a), the deseasonalised SBUV total column ozone is also given (orange), with the regression model (red) and the non-linear trend (thick, red). Data are shifted so that the trend line is zero in 1998. DLM results for WACCM-SD (blue) and SOCOL-SD (purple) from Fig. S11 are also shown; model results in (a) are for the stratospheric column.



Geophysical Research Letters



RESEARCH LETTER

10.1029/2018GL078071

Key Points:

- Observations show that lower stratospheric ozone at extrapolar latitudes increased strongly in 2017 relative to a negative anomaly in 2016
- Model simulations reproduce the observed ozone variations well, and the main driver in the lower stratosphere is atmospheric dynamics
- The contribution of an observation-based trend in short-lived chlorine species to recent lower stratospheric ozone variations is small

On the Cause of Recent Variations in Lower Stratospheric Ozone

Martyn P. Chipperfield^{1,2} , Sandip Dhomse¹ , Ryan Hossaini³ , Wuhu Feng^{1,4} ,
Michelle L. Santee⁵ , Mark Weber⁶ , John P. Burrows⁶ , Jeanette D. Wild^{7,8} ,
Diego Loyola⁹ , and Melanie Coldewey-Egbers⁹ 

¹School of Earth and Environment, University of Leeds, Leeds, UK, ²National Centre for Earth Observation, University of Leeds, Leeds, UK, ³Lancaster Environment Centre, Lancaster University, Lancaster, UK, ⁴National Centre for Atmospheric Science, University of Leeds, Leeds, UK, ⁵Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA, ⁶Institute of Environmental Physics, University of Bremen, Bremen, Germany, ⁷Innovim LLC, Greenbelt, MD, USA, ⁸NOAA/NCEP/Climate Prediction Center, College Park, MD, USA, ⁹Deutsches Zentrum für Luft-und Raumfahrt (DLR), Institut für Methodik der Fernerkundung (IMF), Oberpfaffenhofen, Germany

Abstract We use height-resolved and total column satellite observations and 3-D chemical transport

Cause of ozone variations

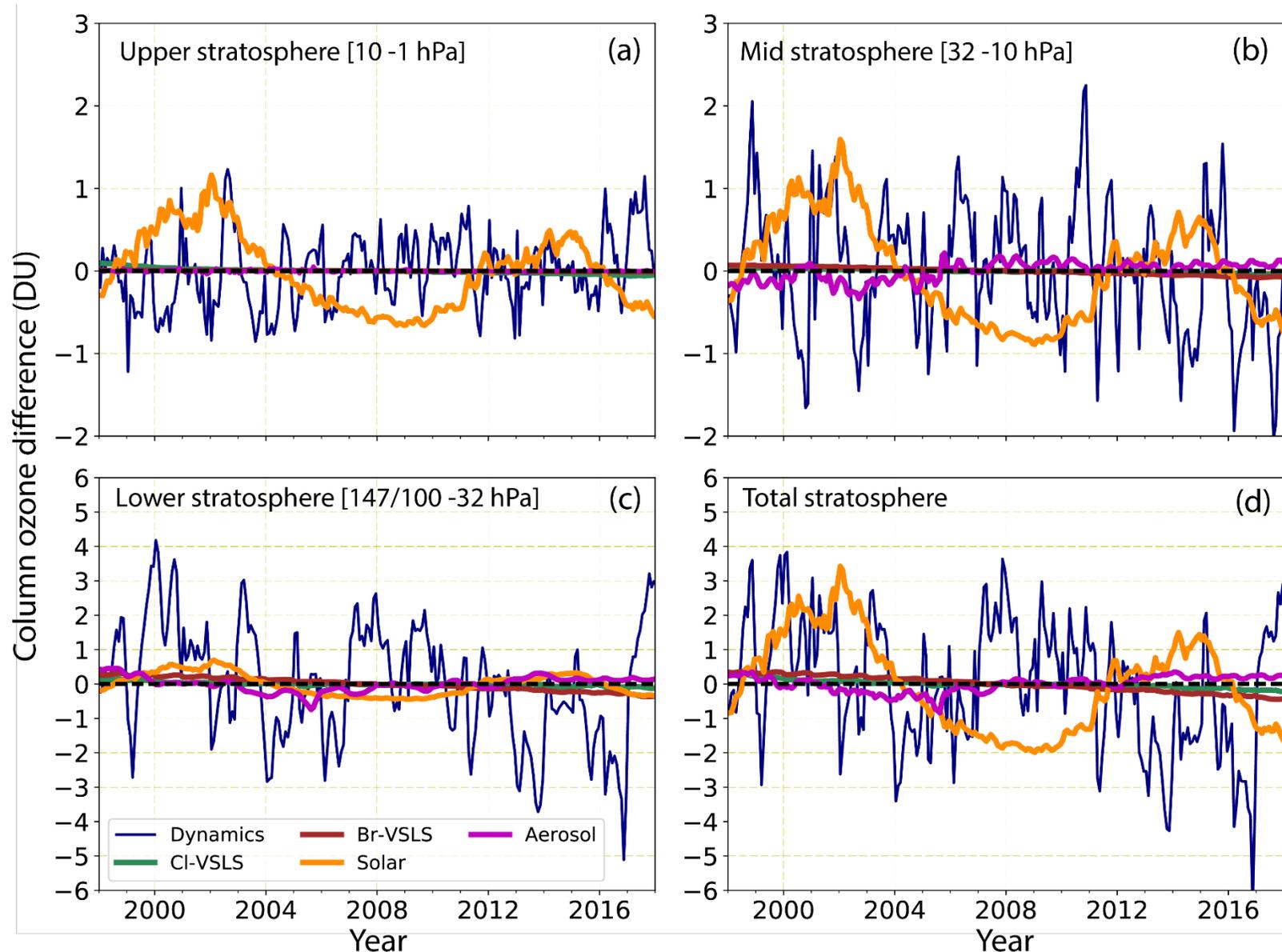


60S – 60N

Model sensitivity experiments.

VSLs effect small (low latitude lower stratosphere).

Dynamics important.



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<https://doi.org/10.5194/acp-18-8409-2018>
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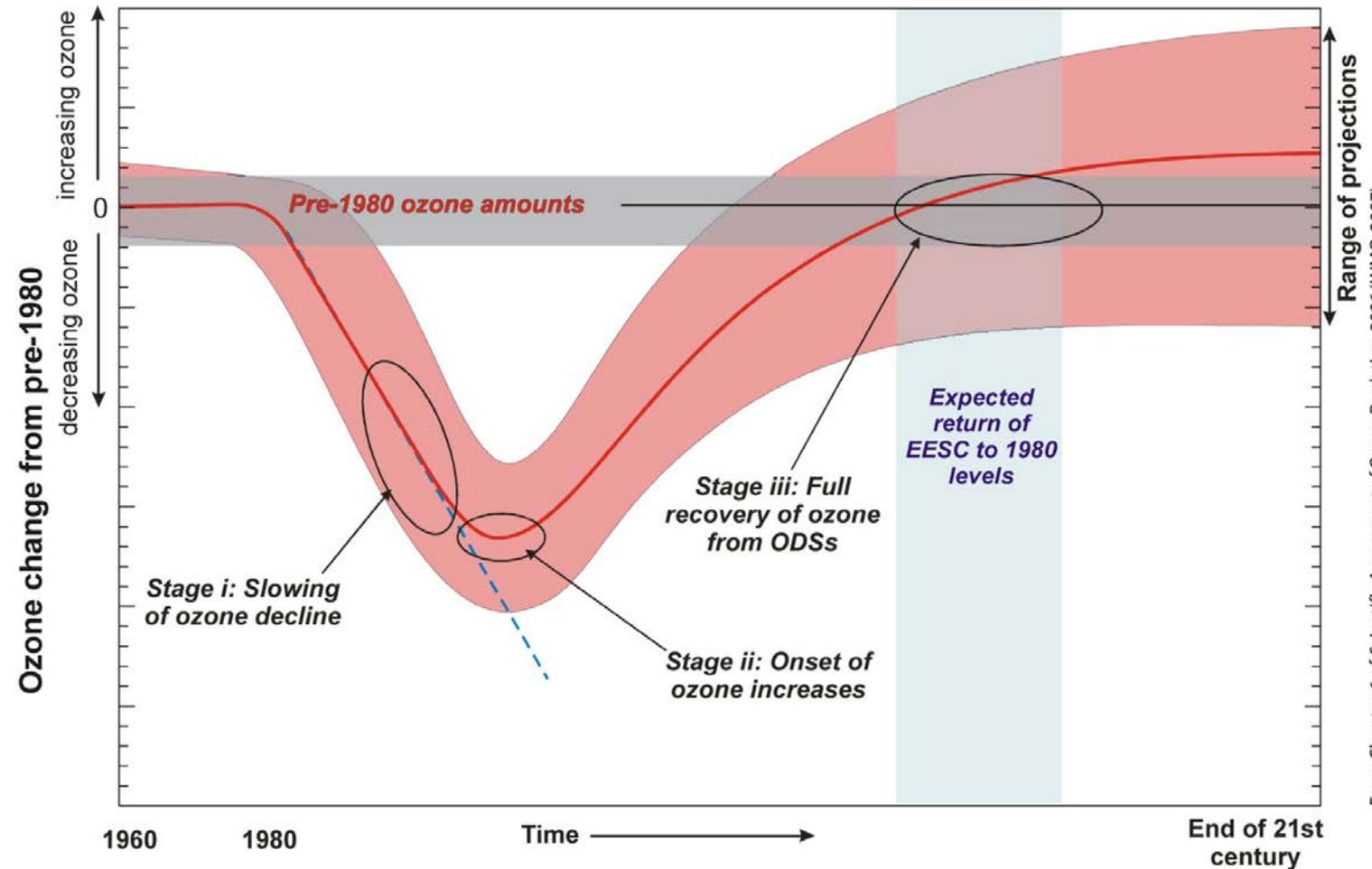


Atmospheric
Chemistry
and Physics
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Estimates of ozone return dates from Chemistry-Climate Model Initiative simulations

Sandip S. Dhomse¹, Douglas Kinnison², Martyn P. Chipperfield^{1,3}, Ross J. Salawitch^{4,5,6}, Irene Cionni⁷, Michaela I. Hegglin⁸, N. Luke Abraham^{9,10}, Hideharu Akiyoshi¹¹, Alex T. Archibald^{9,10}, Ewa M. Bednarz⁹, Slimane Bekki¹², Peter Braesicke¹³, Neal Butchart¹⁴, Martin Dameris¹⁵, Makoto Deushi¹⁶, Stacey Frith^{17,18}, Steven C. Hardiman¹⁴, Birgit Hassler¹⁵, Larry W. Horowitz¹⁹, Rong-Ming Hu¹², Patrick Jöckel¹⁵, Beatrice Josse²⁰, Oliver Kirner²¹, Stefanie Kremser²², Ulrike Langematz²³, Jared Lewis²², Marion Marchand¹², Meiyun Lin^{19,24}, Eva Mancini²⁵, Virginie Marécal²⁰, Martine Michou²⁰, Olaf Morgenstern²⁶, Fiona M. O'Connor¹⁴, Luke Oman¹⁸, Giovanni Pitari²⁷, David A. Plummer²⁸, John A. Pyle^{9,10}, Laura E. Revell^{22,29}, Eugene Rozanov^{29,30}, Robyn Schofield^{31,32}, Andrea Stenke²⁹, Kane Stone^{31,32,a}, Kengo Sudo^{33,34}, Simone Tilmes², Daniele Visioni²⁵, Yousuke Yamashita^{11,34}, and Guang Zeng²⁶

1980 used as reference baseline, but ozone depletion from chlorine and bromine did occur before this time.

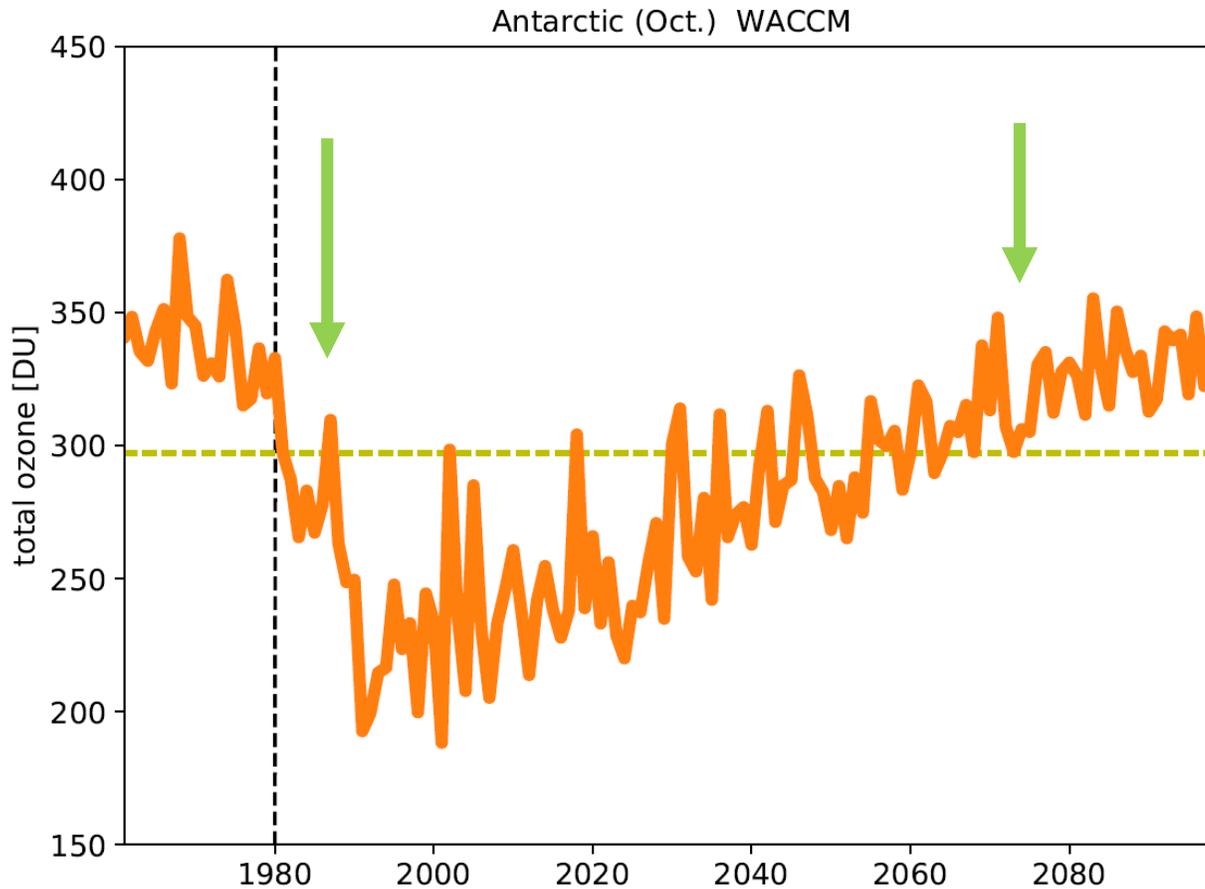


From Chapter 6 of Scientific Assessment of Ozone Depletion: 2006 (WMO, 2007).

What Does a 'Return Date' mean?



Antarctic October mean total column ozone



One realisation of REF-C2 CCMI simulation from WACCM CCM.

Large interannual variability (which may be under/overestimated by the model).

Large spread between:
'first year to show 1980 values'
and
'last year to show 1980 values'.

We need to average and smooth results to get a useful estimate of year (with range) when ozone will return (in an average sense).

Model name	REF-C1	REF-C1SD	REF-C2	SEN-C2-FGHG	SEN-C2-FODS	SEN-C2-RCP20	SEN-C2-RCP45	SEN-C2-RCP85	SEN-C2-fCH4	SEN-C2-CH4RCP85	SEN-C2-fN2O	Total simulation
ACCESS CCM	1		2	2	2							7
CCSRNIES MIROC3.2	3	1	2	1	1	1	1	1	1		1	13
CESM1 CAM4-CHEM	3		3									6
CESM1 WACCM	5	1	3	3	3		1	3	1	1	1	22
CHASER (MIROC-ESM)		1	1	1	1	1		1	1		1	8
CMAM	3	1	1	1	1	1	1	1	1	1	1	13
CNRM-CM5-3	4	1	2									7
EMAC-L47	1	1	1									3
EMAC-L90	1	1	1									3
GEOSCCM	1		1						1	1	1	5
GFDL-CM3/AM3	1	1	1				3	1				7
HadGEM3-ES	1	1	1									3
IPSL-LMDZ-REPROBUS	1	1	1	1	1							3
MRI	1	1	1									3
NIWA-UKCA	3		5	3	2				1		1	15
SOCOL3	3		1			1	1	1	1	1	1	10
TOMCAT (CTM)	1	1										2
ULAQ CCM	3		3	1	1	1	1	1	1	1	1	14
UMSLIMCAT	1		1	1	1		1	1		1		7
UMUKCA-UCAM	1	1	2									4
Total realisations	38	13	33	13	12	5	9	10	8	6	8	155
Total models	19	13	19	8	8	5	7	8	8	6	8	

20 models
155 realisations

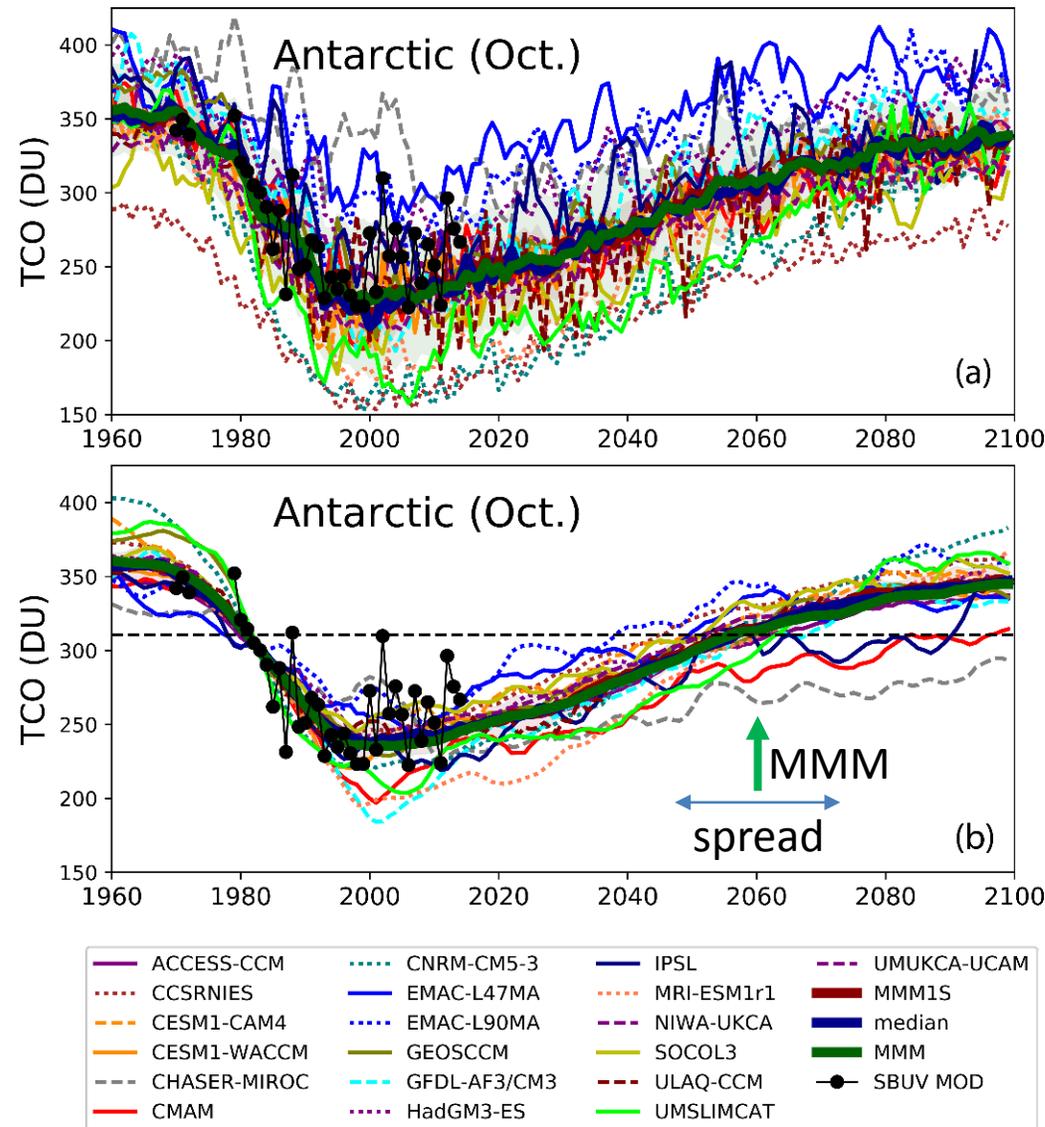
‘Core’ simulations well represented.

REF-C2 – Main simulation (RCP 6.0)

Patchy coverage of ‘sensitivity’ simulations
(fixed ODS, GHG, climate, N₂O, CH₄).

Limits ability to extract robust results from sensitivity runs.

Example for October
Antarctic total
column ozone



Results from 19 models
- Mean of ensemble where relevant.
- Otherwise 3-year smoothing.

Model results adjusted to fit observed column in 1980.

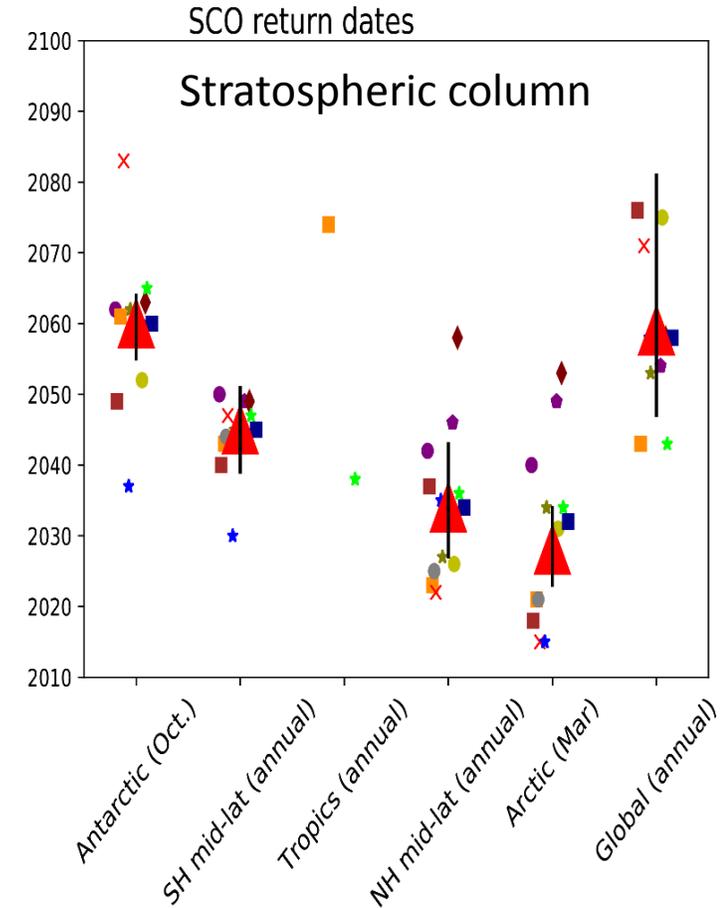
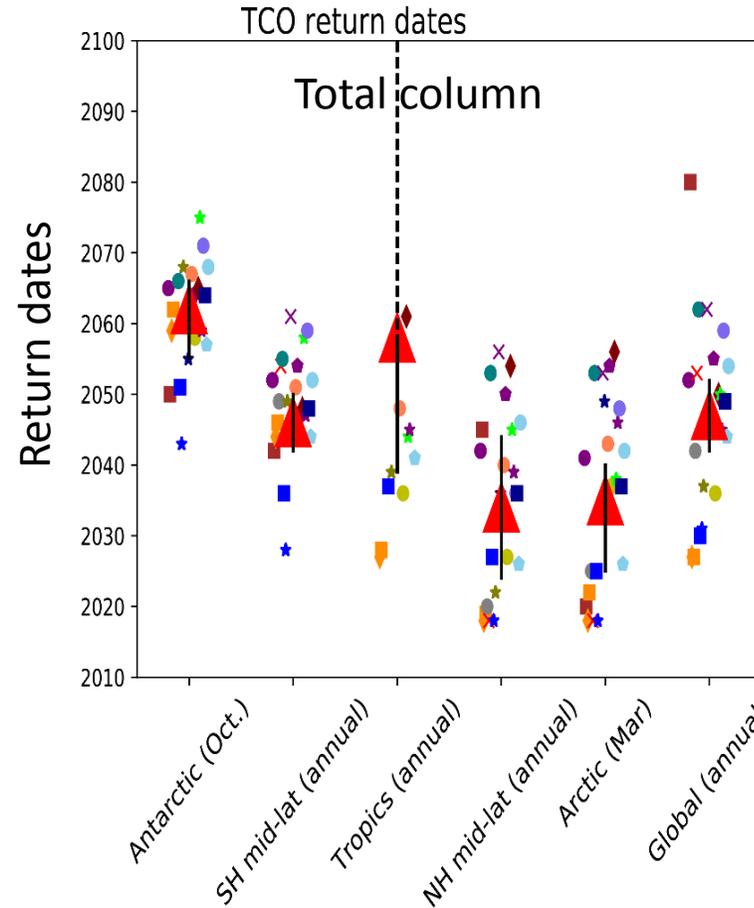
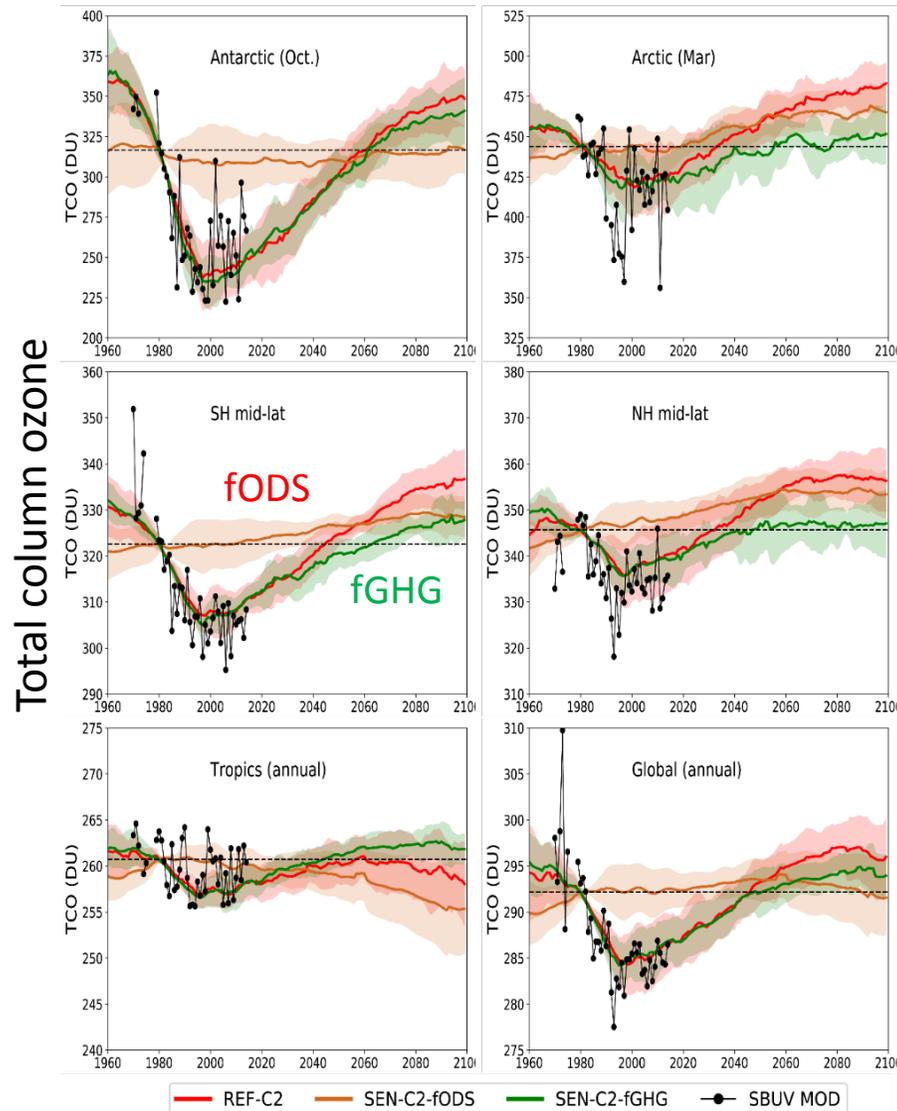
Additional smoothing

'MMM1S' used to estimate return dates.

Return Dates in Different Latitude Bands



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- | | | | |
|----------------|--------------|----------------|---------------|
| ● ACCESS-CCM | × CMAM | ■ GFDL-AF3/CM3 | ● SOCOL3 |
| ■ CCSRNIIES | ● CNRM-CM5-3 | × HadGM3-ES | ◆ ULAQ-CCM |
| ◆ CESM1-CAM4 | ■ EMAC-L47MA | ● MRI-ESM1r1 | ★ UMSLIMCAT |
| ■ CESM1-WACCM | ★ EMAC-L90MA | ◆ NIWA-UKCA | ★ UMUCCA-UCAM |
| ● CHASER-MIROC | ★ GEOSCCM | | |

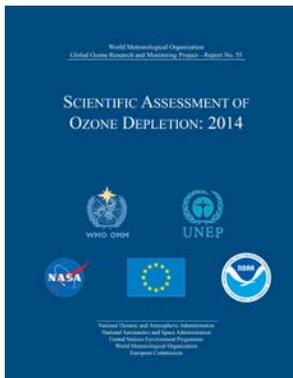
Summary of Return Dates



Model Scenario	WMO	CMIP5	CCMI REF-C2	CCMI 19 models RCP 6.0
	(2011, 2014)	Eyring et al. (2013)	Multi Model Mean 1 sigma MMM1S	
SH pole (October)	2050 (2045-2060)	2046 (2040-2055)	2062 (2055-2066)	
SH Mid-latitudes	2035 (2030-2040)	2041 (2033-2046)	2046 (2042-2050)	
Tropics	2042 (2028-)	N/A	2058 (2039-2100)	
NH Mid-latitudes	2021 (2017-2026)	2032 (2026-2039)	2034 (2024-2044)	
NH pole (March)	2030 (2025-2035)	2028 (2020-2033)	2035 (2025-2040)	
Global (60°S-60°N)	2032 (2027-2038)	2043 (2035-2050)	2047 (2042-2052)	

CCMVal-2 models
A1b GHG scenario

CMIP5
5 models
RCP 6.0



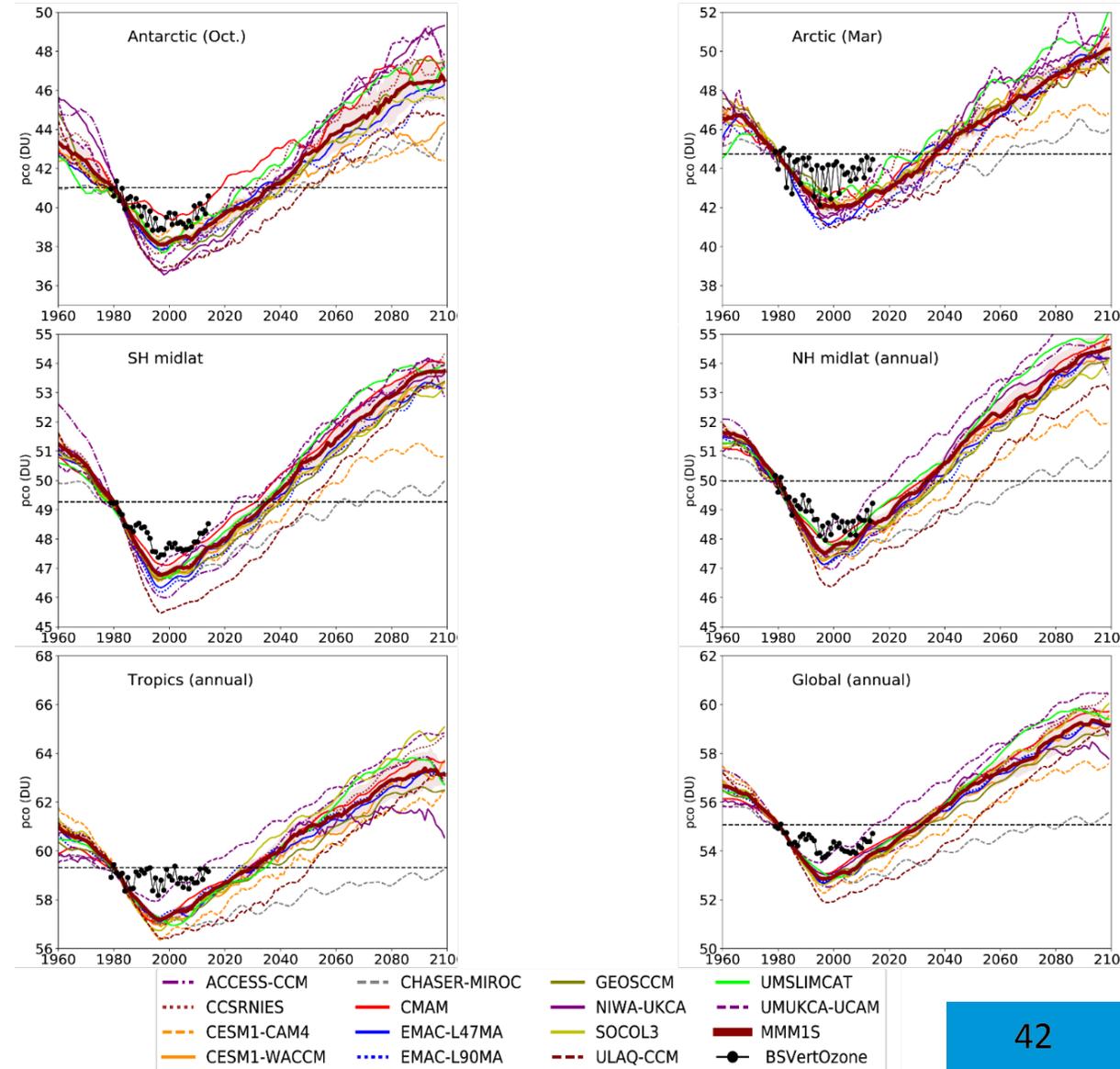
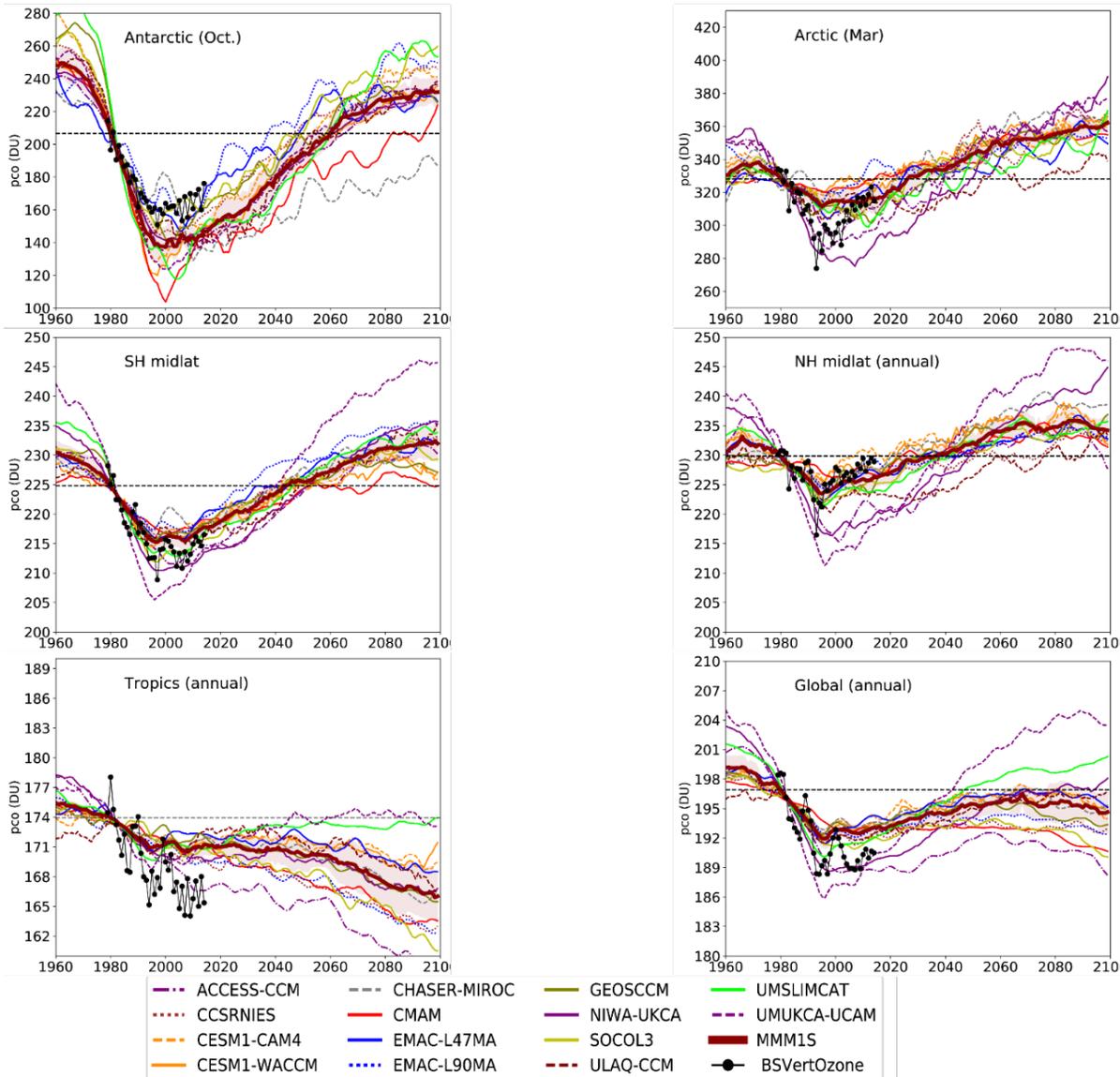
Lower versus Upper Stratosphere



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Lower stratosphere

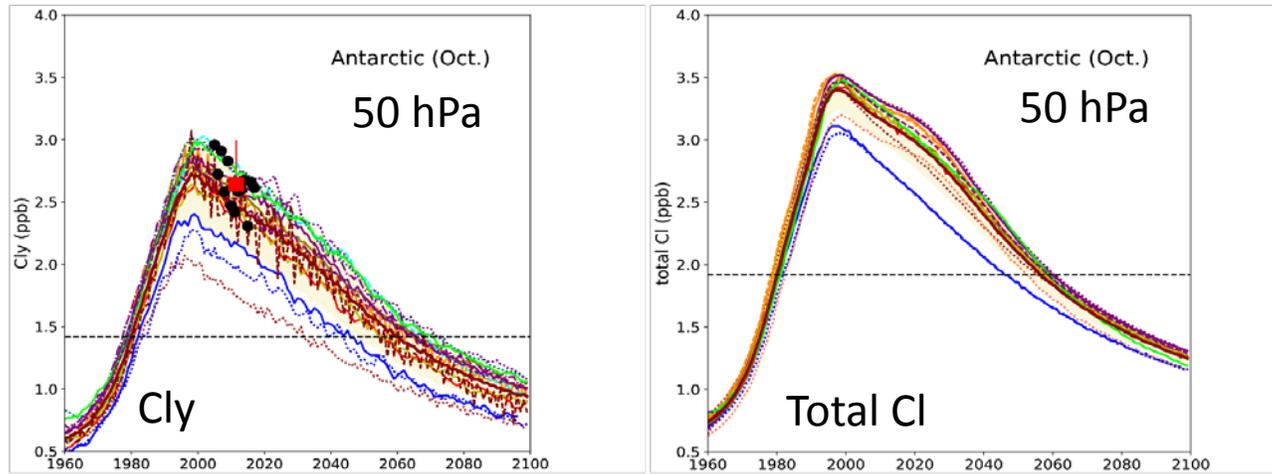
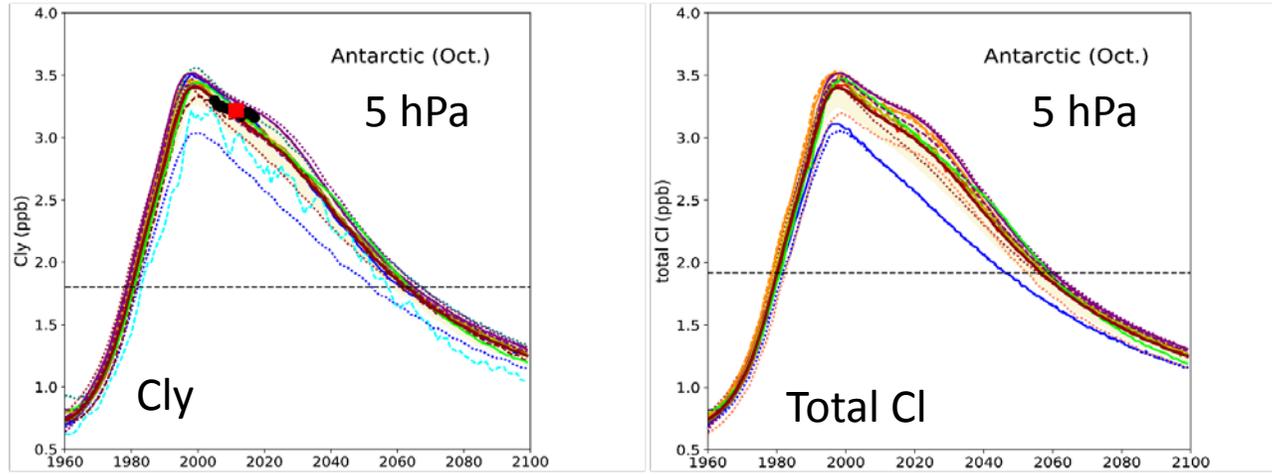
Upper stratosphere



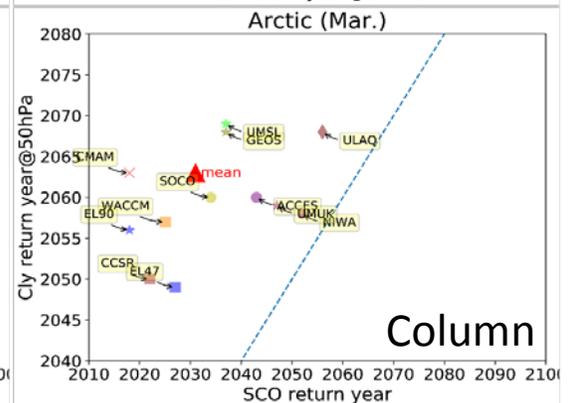
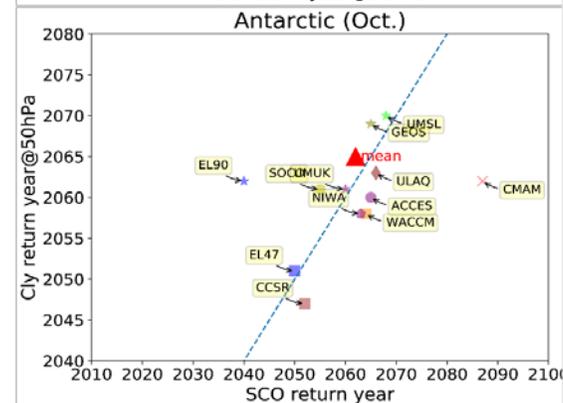
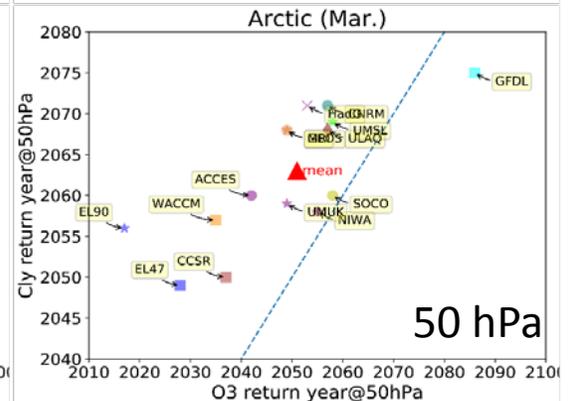
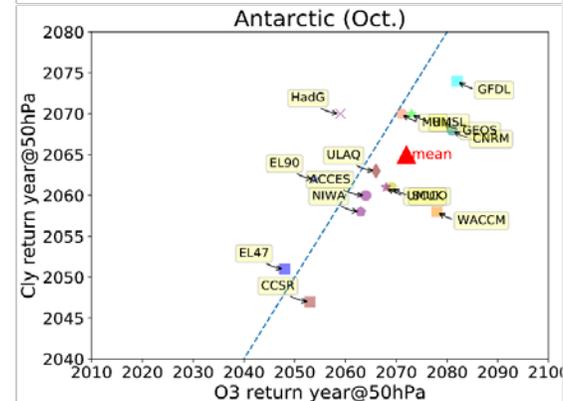
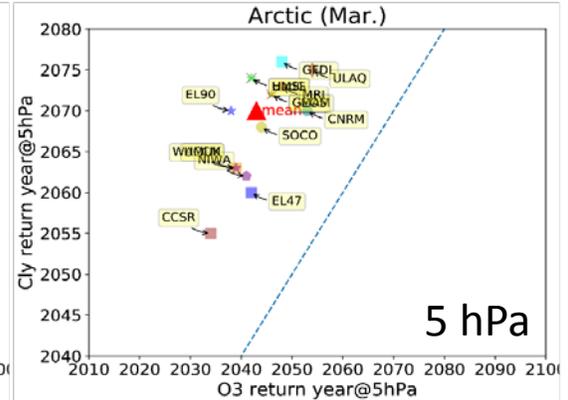
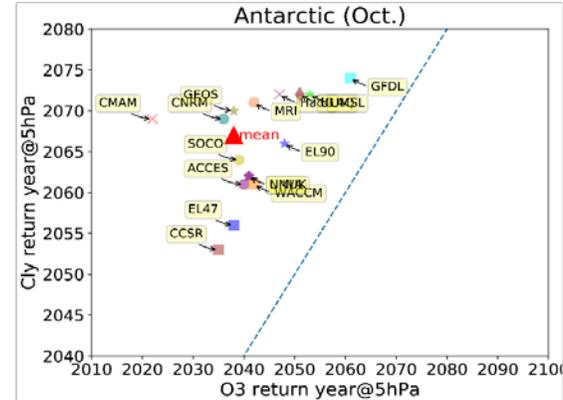
Correlation with Chlorine Loading



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|-------------|------------|--------------|-------------|-----------------|
| ACCESS-CCM | CNRM-CM5-3 | GFDL-AF3/CM3 | ULAQ-CCM | MMM1S |
| CCSRNIES | EMAC-L47MA | HadGEM3-ES | UMSLIMCAT | ● MLS (HCl+ClO) |
| CESM1-WACCM | EMAC-L90MA | NIWA-UKCA | UMUKCA-UCAM | ■ MLS mean |
| CMAM | GEOSCCM | SOCOL3 | | |



- Ozone layer has started to 'recover' in some regions, but at limit of detection in others.
- Montreal Protocol has already had significant benefits and is working.
- Some anthropogenic chlorine VSLs are increasing. Impact so far likely small but sources need to be understood and monitored.
- Evidence for some decrease in lower stratosphere ozone in recent years. Large interannual variability (not all trend). Due to dynamics?
- CCMs can predict ozone return dates. Largely controlled by halogen return dates, modulated by climate change (T, dynamics).

