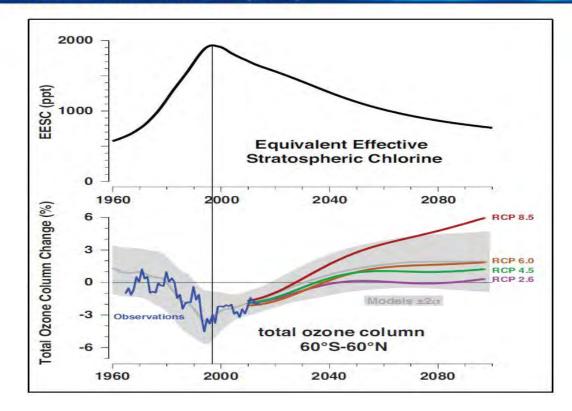
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Ozone variability and long-term changes Michel Van Roozendael, BIRA-IASB

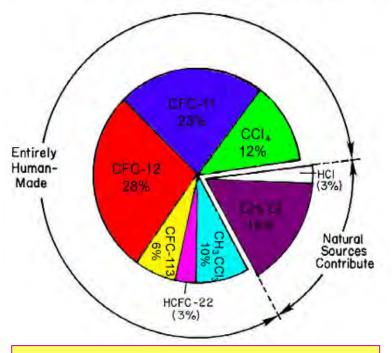


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- 1928: start of CFC production
- 1971: 1st observation of CFC in the atmosphere (J. Lovelock)
- 1974: identification of O₃ destruction potential by CFC: Rowland & Molina
- 1995: Nobel prize in chemistry: F. Rowland, M. Molina & P. Crutzen, 1995



Primary Sources Of Chlorine Entering The Stratosphere



1990: 80% of stratospheric chlorine is of anthropogenic origin

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Long term ozone depletion

- Increasing anthropogenic emissions, since industrial revolution, esp. CFCs since 1960
 increasing concentrations of destructive radicals
 - \clubsuit steady decrease of O_3 on a global scale
- Recurrent spring-time ozone hole at the South pole
- Seasonal ozone depletion in the North pole, strongly modulated by dynamics

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Nature, Vol. 315, 16 May 1985

Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction

J. C. Farman, B. G. Gardiner & J. D. Shanklin

British Antarctic Survey, Natural Environment Research Council, High Cross, Madingley Road, Cambridge CB3 0ET, UK



 First alarming signs of O₃ degradation were given in 1984 and 1985 based on

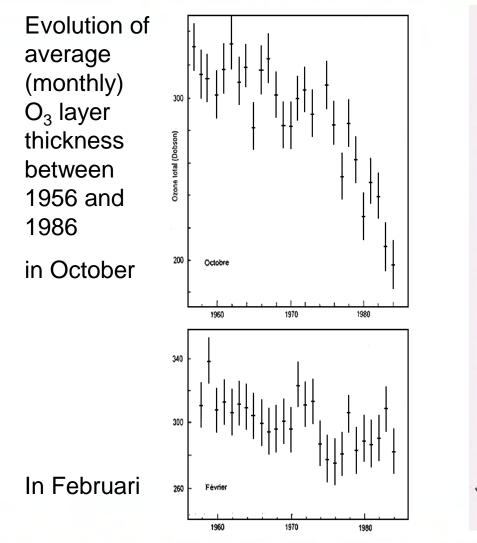
> easurements at I Halley Bay 7)

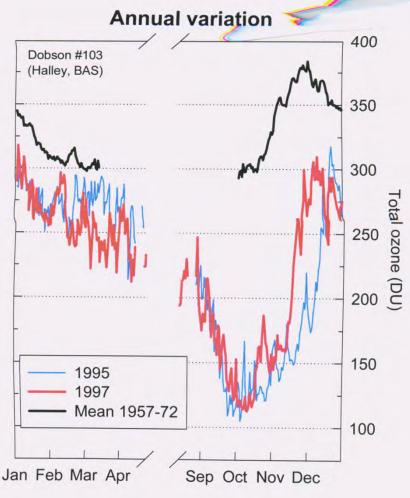
er thins lly in the local vn to ~ 100 DU

Dr. Shigeru Chubachi at Ozone Commission meeting in Halkidiki (Sept 1984)

nenon was not

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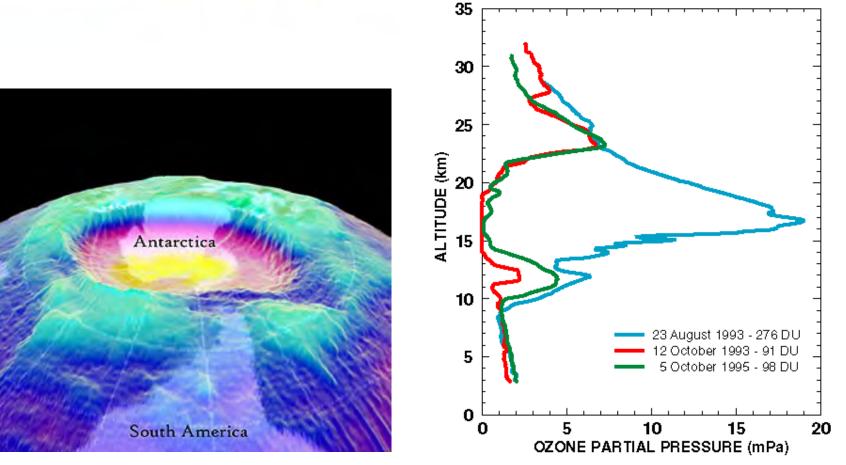
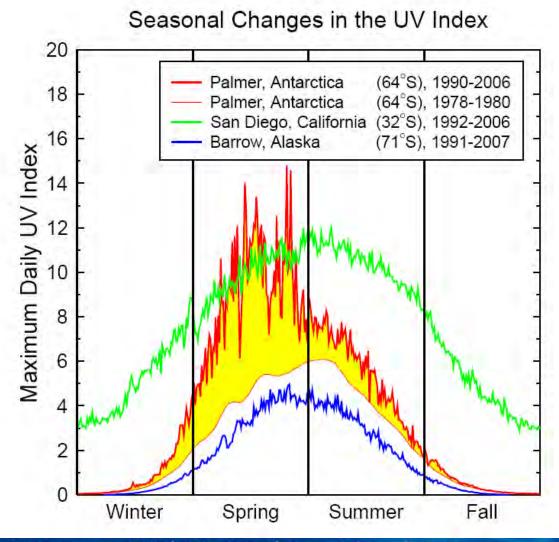


FIG. 16 Ozone profile (partial pressure, mPa) measured by balloon-borne ozonesonde at the South Pole on 5 October 1995 (green line), and comparison to profiles measured in 1993. (Source: CPC)

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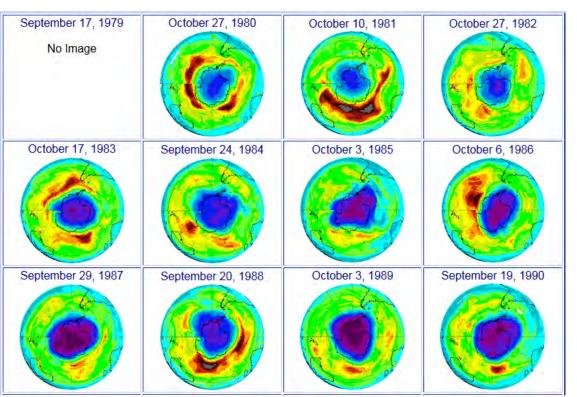
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When the first TOMS measurements were taken the drop in ozone levels in the stratosphere was so dramatic that at first the scientists thought their instruments were faulty...

TOMS quickly confirmed the results from Farman et al., and the term Antarctic ozone hole entered popular language. NASA Nimbus 7



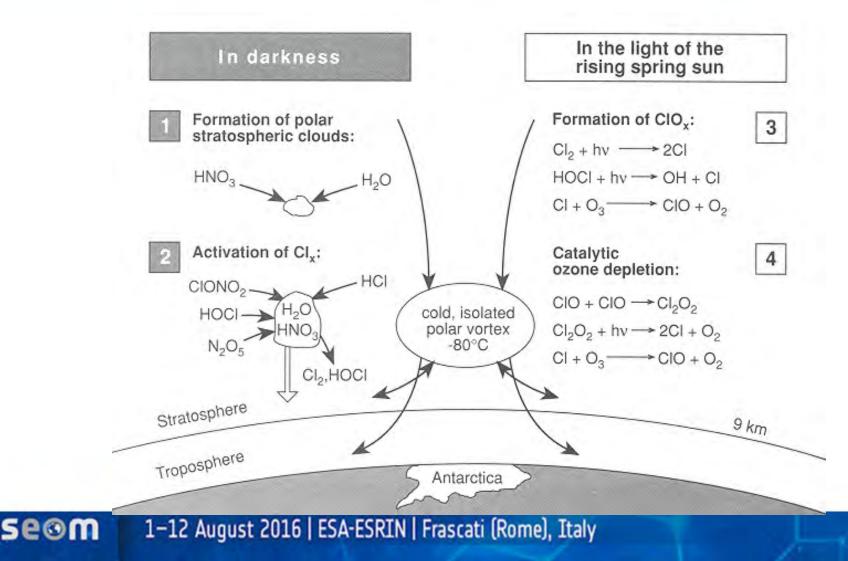






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Heterogeneous chemistry on PSCs



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Ozone chemistry in polar regions

Main catalytic cycles leading to the formation of the ozone hole

```
2 (Cl + O_3 \rightarrow ClO + O_2)

2 ClO + M \rightarrow Cl_2O_2 + M

Cl_2O_2 + hv \rightarrow Cl + ClOO

ClOO + M \rightarrow Cl + O_2 + M
```

Net: 2 $O_{3+}h\upsilon \rightarrow 3 O_{2}$

- require light !
- require the presence of chlorine and bromine molecules in sufficient amounts

70%

 $\begin{array}{l} Br + O_3 \rightarrow BrO + O_2 \\ CI + O_3 \rightarrow CIO + O_2 \\ BrO + CIO \rightarrow Br + CI + O_2 \\ \end{array}$ Net: 2 $O_3 \rightarrow 3 O_2$

30%



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Ozone Abundance

Experimental evidence

Ozone and reactive chlorine measurements from a flight through the ozone hole in Antarctica, 1987 3000 1.5 Ozone 2500 (Scale at Left) orine Abundance 1.0 2000 (Parts per Billion) per Billion 1500 1200 3000 CIO MIXING RATIO (ppt) (a) 23 August 16 September O₃ MIXING RATIO (ppb) 1000 CIO 500 Reactive Chlorine 0, (Scale at Right) 600 2000 0 CIO 1000 0 68 70 72 68 70 72 66 62 64 66 62 64 LATITUDE (°S) 2042:8.17:1/96:blm



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Examples of ozone depleting source gases (ODS)

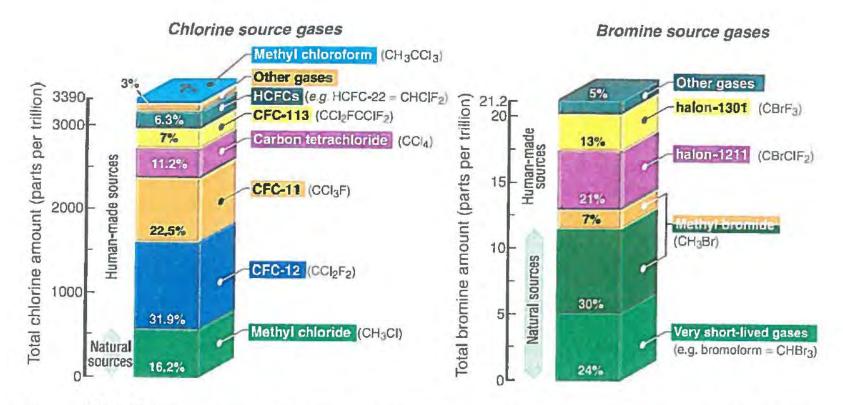


Figure 11.5 Primary sources of chlorine and bromine transported to the stratosphere in 2004. Source: Scientific Assessment of Ozone Depletion: 2006, World Meteorological Organization, Global Ozone Research and Monitoring Project, Report No. 50, WMO, 2007.

Situation 2004



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Potential	Atmospheric lifetime (years)	Steady-state ozone depletion	
trace gas		Model	Semiempirical
CFC-11	50	1.0	1.0
CFC-12	102	0.82	0.9
CFC-113	85	0.90	0.9
CH ₃ CCl ₃	5.4	0.12	0.12
HCFC-22	13.3	0.04	0,05
HCFC-123	1.4	0.014	0.02
HCFC-141b	9.4	0.10	0.1
HCFC-142b	19.5	0.05	0.066
HFC-134a	14	$< 1.5 \times 10^{-5}$	$<5 \times 10^{-4}$
HFC-125	36	$<3 \times 10^{-5}$	
CH ₃ Br	$1.3(0.7)^d$	0.64	$0.57(0.39)^d$
H-1301		12	13
H-1211		5.1	5
CH ₂ ClBr	$0.23 - 0.36^{c}$	$0.098 - 0.15^{c}$	
CH ₂ BrCH ₂ CH ₃	0.029 ^c	0.026 ^c	

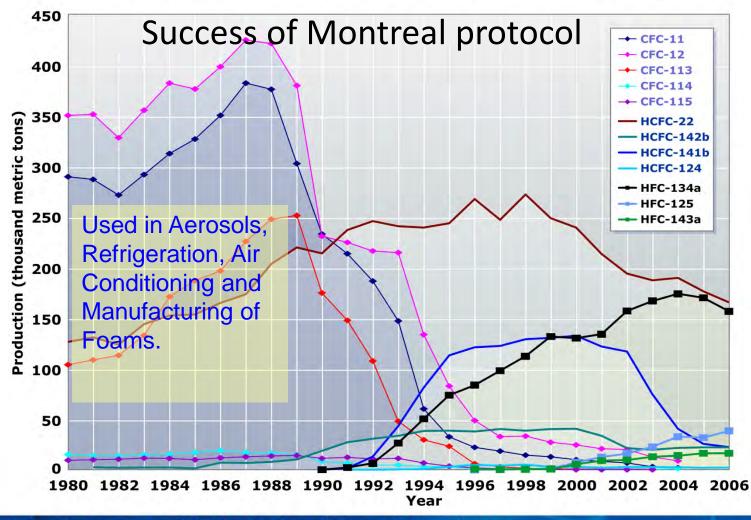
 TABLE 13.3
 Atmospheric Lifetimes and Steady-State Ozone Depletion Potentials (ODP)

 Predicted Using Either a Two-Dimensional Model or a Semiempirical Method^{a,b}

1981	Policies	Scientific Assessments C The Stratosphere 1981. Theory and Measurements. WMO No. 11 C	
1901			
1985	Vienna Convention	Atmospheric Ozone 1985. WMO No.16.	
	Montreal Protocol		
1988		International Ozone Trends Panel Report 1988.	
1000		Two volumes. WMO No. 18	
1989		Scientific Assessment of Stratospheric Ozone: 1989.	
1000	London Adjustments on	Two volumes. WMO No. 20.	
1990	London Adjustments an		
1990		Climate Change, The IPCC first Scientific Assessment, Impacts Assessment and Response Strategies Reports	
1991		Scientific Assessment of Ozone Depletion: 1991.	
1771		WMO No. 25.	
1992		Methyl Bromide: Its Atmospheric Science, Technology, and Economics (Assessment Supplement).	
1//2		UNEP (1992).	
1992	Copenhagen Adjustmen		
1992			
		IPCC Supplementary Report to the Scientific Assessment	
1994		Scientific Assessment of Ozone Depletion: 1994. WMO No. 37	
		Climate Change 1994, Radiative Forcing of Climate Change and an Evaluation of the IPCC IS92	
		Emission Scenarios	
1995	Vienna Adjustment		
1995	-	Climate Change 1995. The IPCC second Scientific Assessment, Impacts Assessment Reports	
1997	Montreal Adjustments and Amendment		
	Kyoto Protocol (UNFCC	CC third session, Kyoto, Dec. 1997)	
1998		Scientific Assessment of Ozone Depletion: 1998. WMO. No. 44	
1999	Beijing (China) Adjustn		
2000		The IPCC third Scientific Assessment, Impacts Assessment Reports	
2002		Scientific Assessment of Ozone Depletion: 2002. WMO. No. 47	
2007		Scientific Assessment of Ozone Depletion: 2006. WMO. No. 50	
2007	th	The IPCC fourth Assessment Report: Climate Change 2007	
2007	Montreal 19 th meeting	of the Parties: 191 countries agree to strengthen protection of the ozone layer by reducing HCFCs	
2008	Doha 20 th meeting of	the Parties: Decision making on destruction ODS and funding phase-out HCFCs	
2011	Scier	ntific Assessment of ozone depletion: 2010. WMO No. 52	
2013		PCC 5th assessment report	
2014		ntific Assessment of ozone depletion: 2014. WMO No. 55	
	Ociei		
3		ust 2016 ESA-ESRIN Frascati (Rome), Italy	

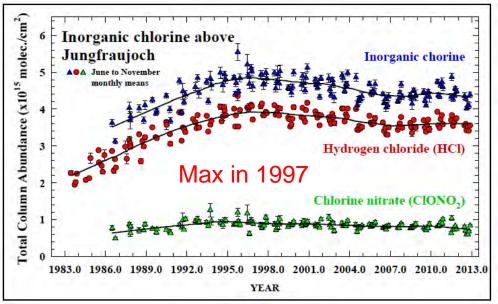
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Annual Production of Fluorocarbons Reported to AFEAS (1980-2006)



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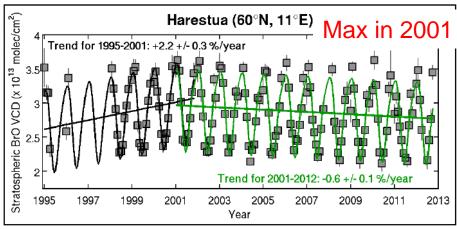
Effect of Montreal Protocol



fits were applied to these data sets (black curves) to provide trends estimates. **E. Mahieu, U. Liège**

Reduction of chlorine and bromine in the stratosphere follows decrease of concentrations of the surface with a delay of 3 to 5 years

= time to reach from troposphere stratosphere.

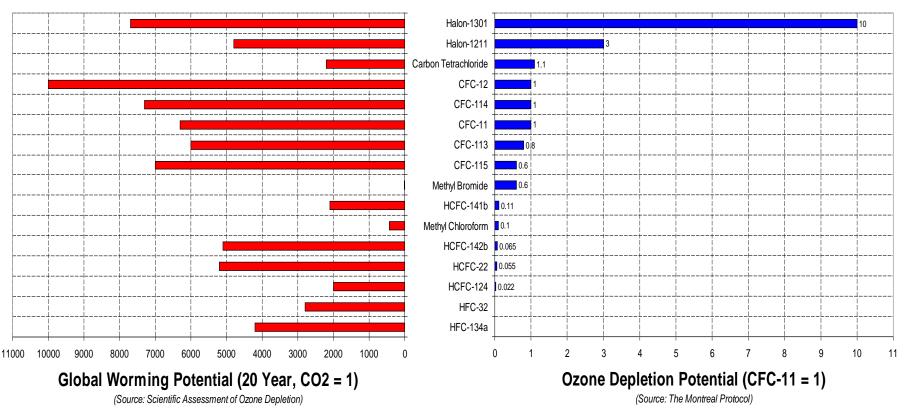


F. Hendrick, IASB



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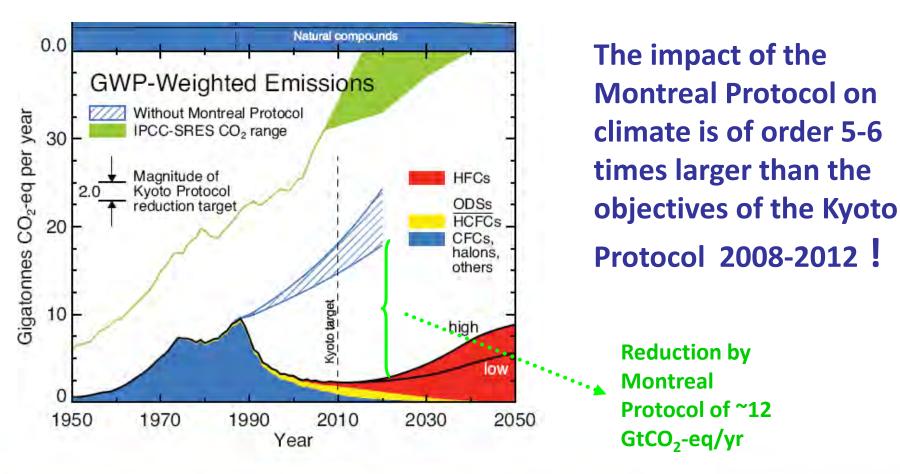
Most ODS are also greenhouse gases!



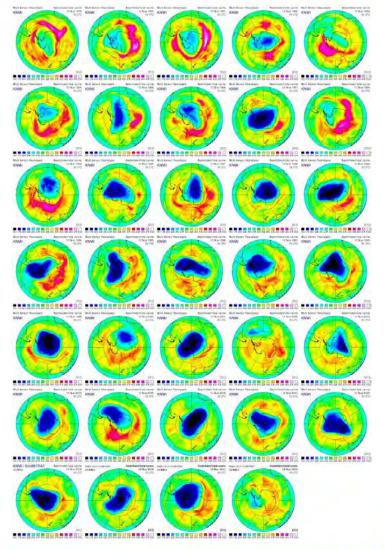
- Montreal Protocol has been beneficial for our climate
- Challenge: search for CFC substitutes that have a negligible GWP !

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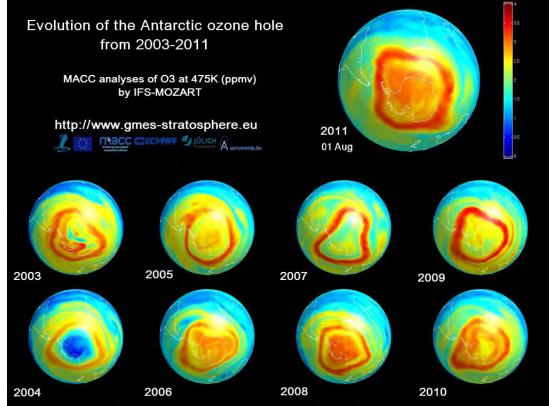
Success of the Montreal Protocol



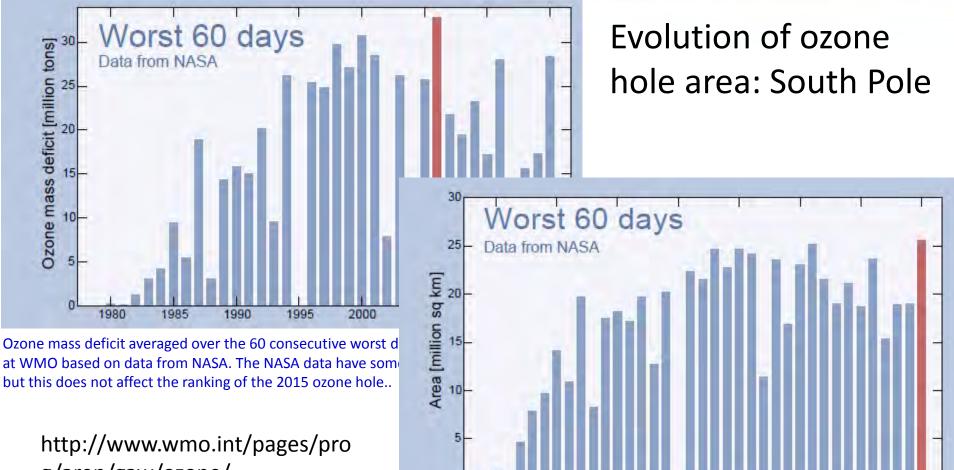
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Monitoring of the ozone hole using satellites and models



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1980

1985

1990

Area of the ozone hole for the years 1979-2015, averaged over the 60 consecutive worst days. The plot is produced at WMO, based on data from NASA. The NASA 1-12 August 2016 | ESA-ES data have some gaps in the mid 1990s, but this does not affect the ranking of the 2015 ozone hole.

1995

2000

2005

2010

2015

at WMO based on data from NASA. The NASA data have some but this does not affect the ranking of the 2015 ozone hole..

g/arep/gaw/ozone/

seom

Cesa

1985 [MSR]

1984 [MSR]

→ EARTH OBSERVATION SUMMER SCHOOL

1981 [MSR]

1980 [MSR]

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1979 [MSR]

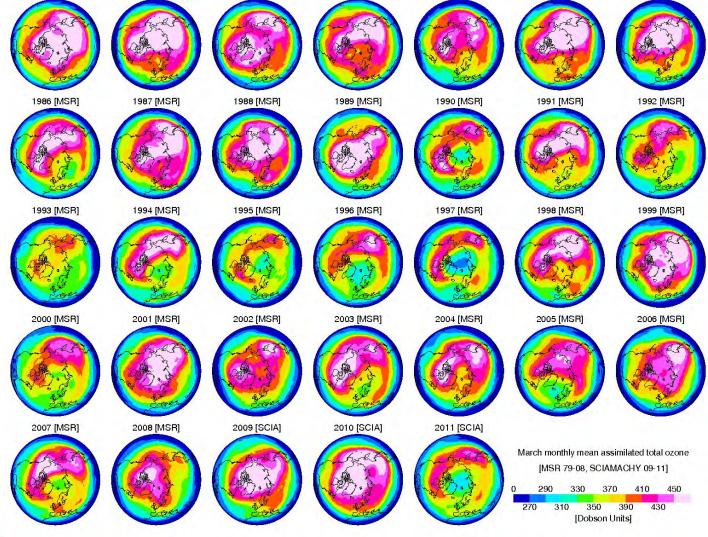
Evolution of the Spring polar ozone over the North Pole from 1979 until 2011

MACC/MSR, KNMI

March monthly

total ozone

mean assimilated



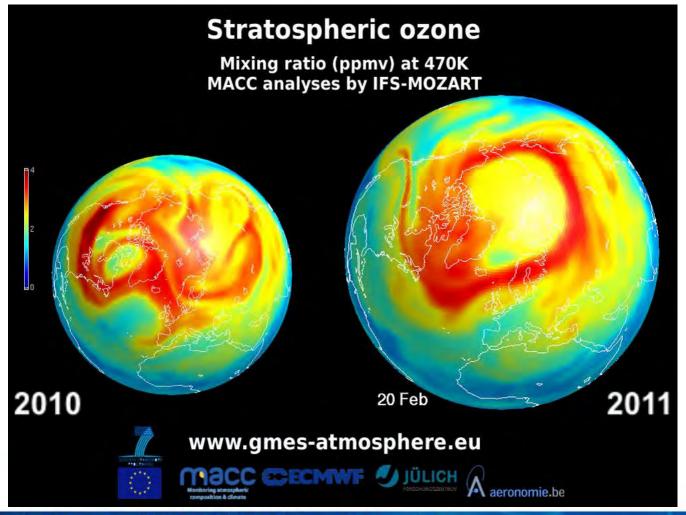
1982 [MSR]

1983 [MSR]



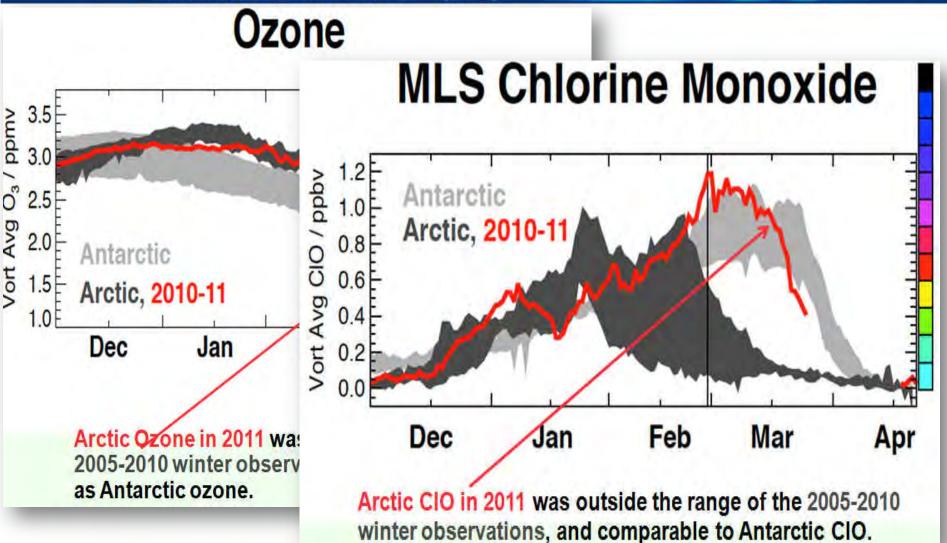
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Most severe Arctic Ozone Hole: 2010-2011





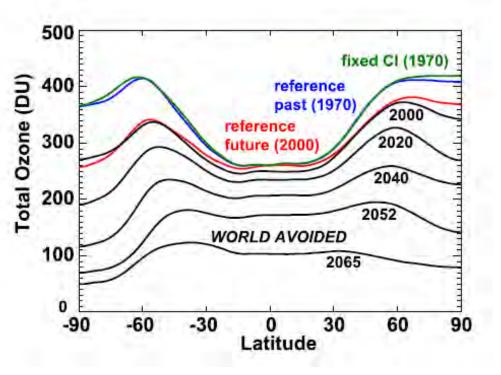
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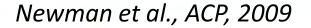




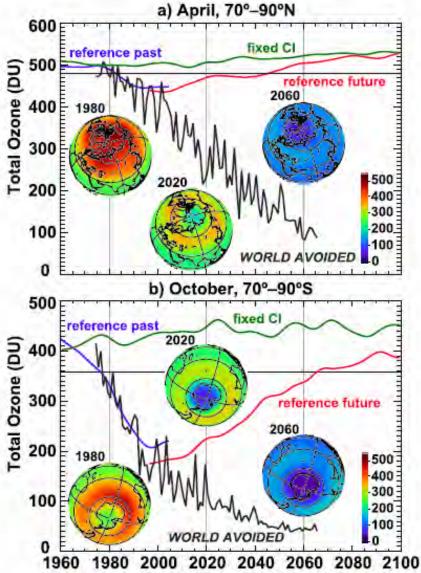
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What would have happened chlorofluorocarbons (CFCs) ha





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Current situation

- O₃ in the middle latitude:
 - 6% (3.5%) lower than average in 1964-1980 SH (NH)
 - UV has not increased since the late 1990s
- O₃ hole at the poles is still as in early 1990 (subject variability) i.e. in Antarctica in October, 40% lower than in 1980
- UV on Antarctic is higher by 55 to 85% than in 1963-1980
- Evidences for changes in SH tropospheric summertime circulation due to Antarctic ozone hole
- The stratosphere has cooled a few K between 1980 and 1995 due to ozone depletion; the cooling reinforced by increasing GHG esp. in recent years
 Ref: WMO 03 Assessment, 2014

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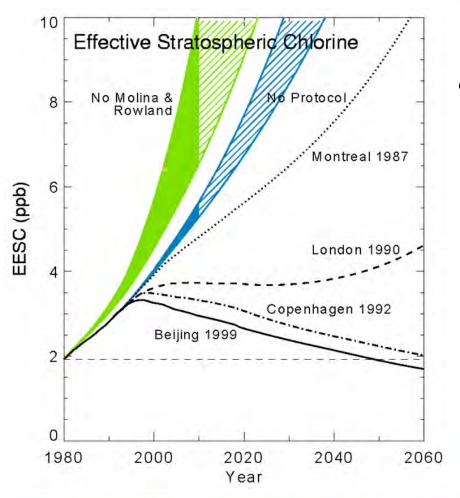
Coupling between ozone and climate

- Concentrations of greenhouse gases (CO₂, CH₄, N₂O,...) rise
- T° rises at the surface; but **decreases** in the stratosphere
- Atmospheric transport changes, rate of chemical reactions changes
- Frequency of formation of clouds, aerosols and PSC change
- Radiation balance changes
- Ozone formation / degradation is influenced, in turn ozone affects UV, T° stratosphere



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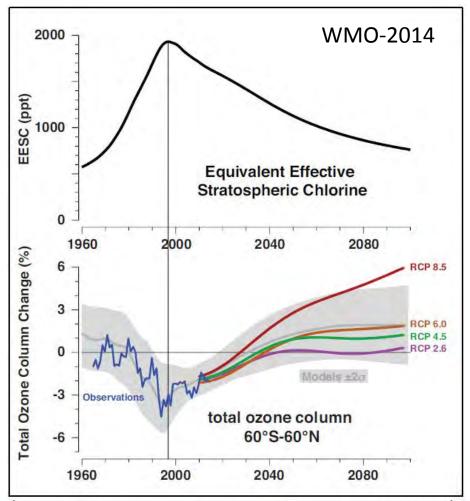
Future evolution of ODS



- EESC back to 1980 levels:
 - ≻ in ~2050
 - ➢ in ~2065 at the poles (older air)

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Future evolution of mid-latitude ozone

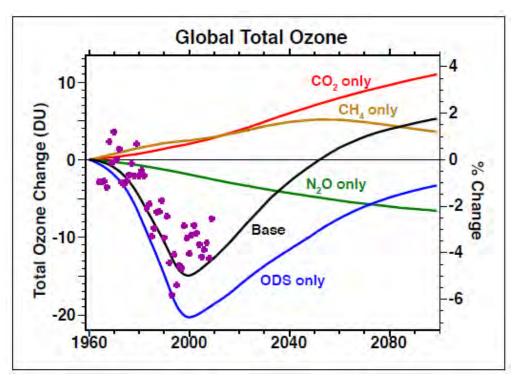


Average total column ozone changes over the same period, from multiple model simulations compared with observations between 1965 and 2013.

Four possible greenhouse gas (CO₂, CH₄, and N₂O) futures are shown. The four scenarios correspond to +2.6 (**purple**), +4.5 (**green**), +6.0 (**brown**), and +8.5 (**red**) W m⁻² of global radiative forcing

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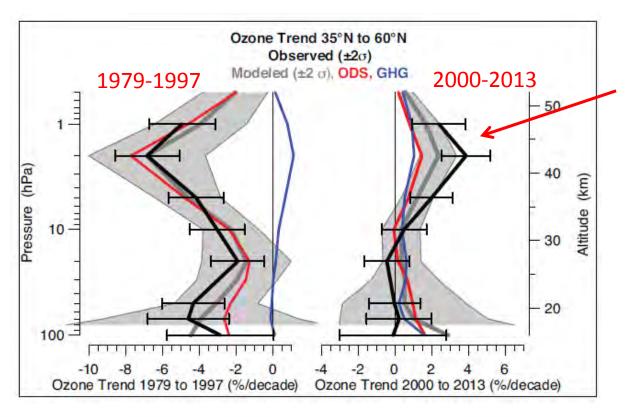
Impact of projected changes in GHGs on ozone



- Increased CO₂, CH₄ and N₂O cools the stratosphere, which tends to increase O₃ because of temperature-dependent chemistry (reduced efficiency of loss)
- Increased CH₄ and N₂O also have further chemical impact on O₃
 - CH₄ increases O₃ by mitigating the effect of halogen-driven O₃ destruction catalytic cycles
 - N₂O decreases O₃ through enhancing the efficiency of the NOx-driven catalytic cycles

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Evidence for O₃ recovery in upper stratosphere



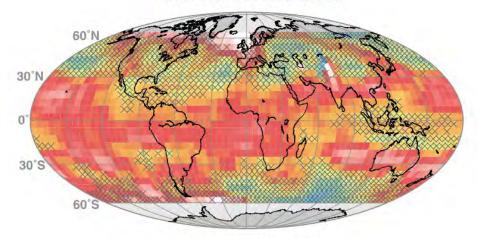
O₃ recovery at 45 km altitude is due to combination of reduction in ODS (Montreal protocol) reinforced by cooling due to GHG increase



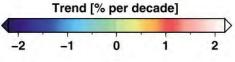
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Regional ozone trend analysis

- Regional trends in total ozone estimated from harmonised multisensor satellite data set (1995-2013)
- Trends in middle latitudes still not significant; recovery masked by natural variability; additional 5-10 years of observations are required



GTO-ECV CCI Total Ozone



Italy

Coldewey-Egbers et al., GRL, 2015

@AGU PUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER 10.1002/2014GL060212

Key Points:

 Global assessment of ozone trends using 18 years of European satellite data
 Natural variability masks ozone recovery in middle latitudes
 Additional 5–10 years of observations are required to detect expected onset

Correspondence to:

A new health check of the ozone layer at global and regional scales

Melanie Coldewey-Egbers¹, Diego G. Loyola R.¹, Peter Braesicke², Martin Dameris³, Michel van Roozendael⁴, Christophe Lerot⁴, and Walter Zimmer¹

¹Remote Sensing Technology Institute, German Aerospace Center, Weßling, Germany, ²Karlsruhe Institute of Technology, Institute for Meteorology and Climate Research, Karlsruhe, Germany, ³Institute for Physics of the Atmosphere, German Aerospace Center, Weßling, Germany, ⁴Belgian Institute for Space Aeronomie BIRA-IASB, Brussels, Belgium

Abstract In this study, we provide a new perspective on the current state of the ozone layer using a comprehensive long-term total ozone data record which has been recently released within the framework.

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3 Fingerprints of Ozone Hole recovery

Science

30 June 2016 release

RESEARCH ARTICLES

Cite as: Solomon et al., Science 10.1126/science.ane0061 (2016).

Emergence of healing in the Antarctic ozone layer

Susan Solomon,1* Diane J. Ivy,1 Doug Kinnison,2 Michael J. Mills,2 Ryan R. Neely III,34 Anja Schmidt3

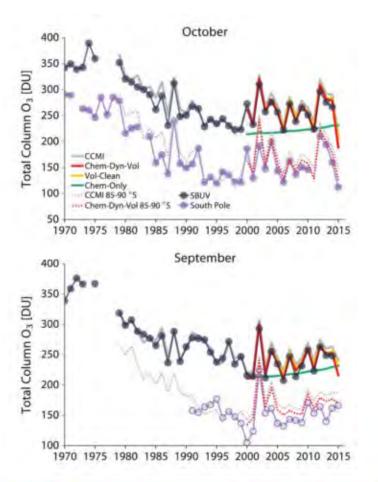
¹Department of Earth, Atmospheric, and Planetary Science, Massachusetts Institute of Technology, Cambridge, MA 02139, USA. ⁴Atmospheric Chemistry Observations and Modeling Laboratory, National Center for Atmospheric Research, PO Box 3000, Boulder, CO 80305, USA. ⁴School of Earth and Environment, University of Leeds, Leeds, UK. ⁴National Centre for Atmospheric Science, University of Leeds, Leeds, UK.

*Corresponding author. Email: solos@mit.edu

Industrial chlorofluorocarbons that cause ozone depletion have been phased out under the Montreal Protocol. A chemically-driven increase in polar ozone (or "healing") is expected in response to this historic agreement. Observations and model calculations taken together indicate that the onset of healing of Antarctic ozone loss has now emerged in September. Fingerprints of September healing since 2000 are identified through (i) increases in ozone column amounts, (ii) changes in the vertical profile of ozone concentration, and (iii) decreases in the areal extent of the ozone hole. Along with chemistry, dynamical and temperature changes contribute to the healing, but could represent feedbacks to chemistry. Volcanic eruptions episodically interfere with healing, particularly during 2015 (when a record October ozone hole occurred following the Calbuco eruption).

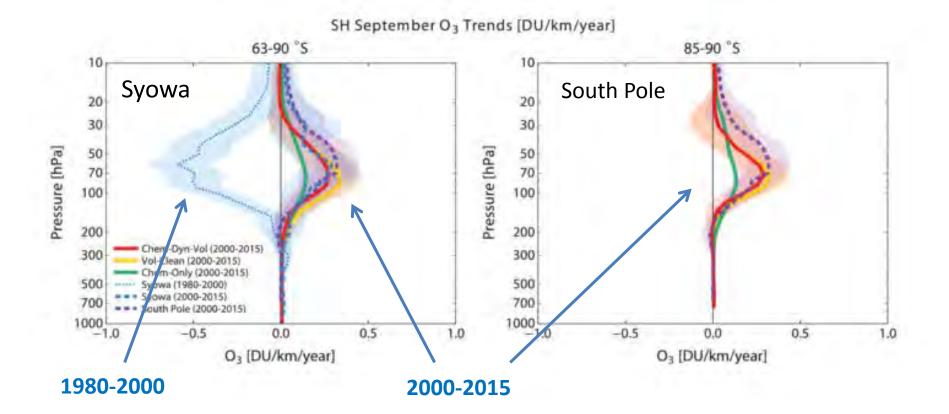
Solomon et al., 2016

(i) increases in ozone column amounts



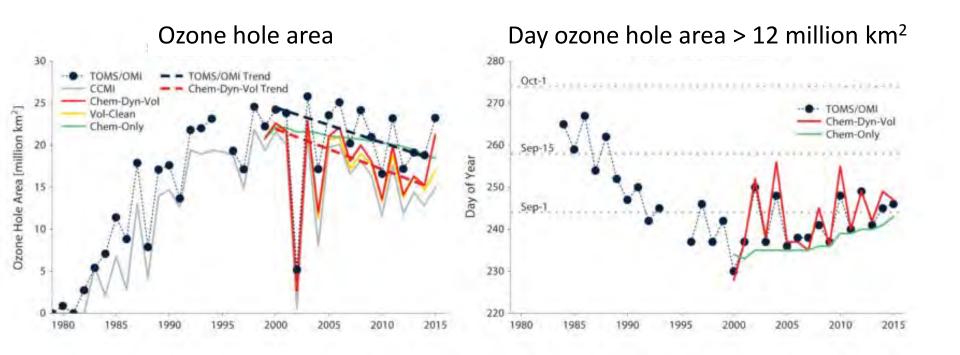
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(ii) changes in the vertical profile of ozone concentration



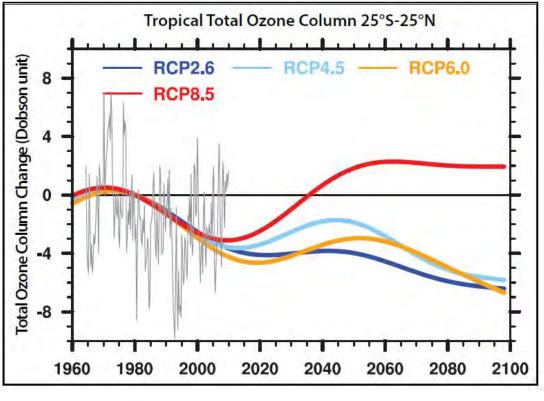
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(iii) decreases in the areal extent of the ozone hole



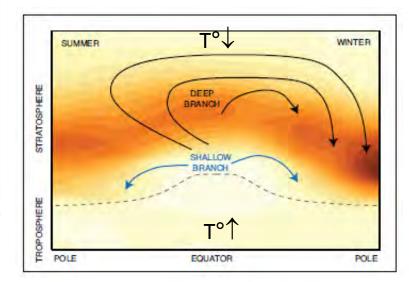
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Predicted reduction of the tropical ozone layer



Recent evidence for changes in tropical upwelling and higher latitudes downwelling due to climate change

 \rightarrow Impact on ozone distribution

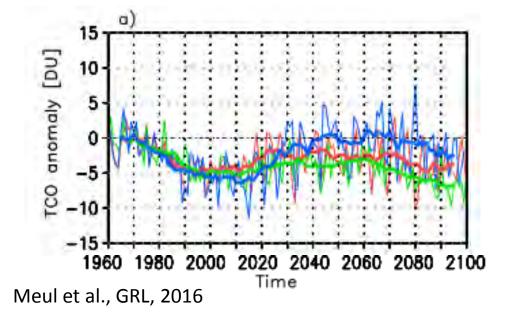


WMO-2014

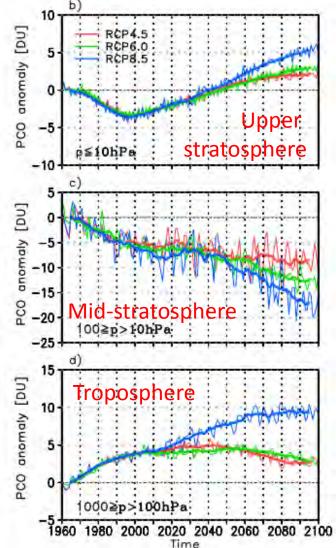


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Impact of rising GHG on tropical ozone



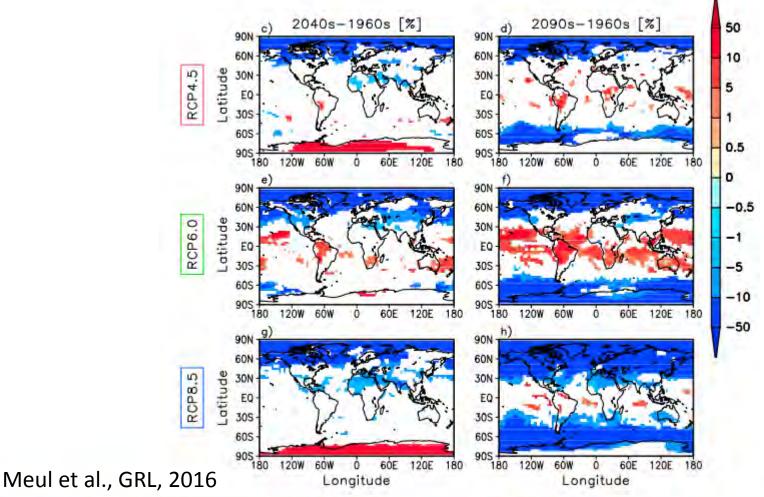
<u>Conclusion</u>: Mid-strat ozone decrease due to change in circulation (partly) compensated by increases to chemical effects at other altitudes





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Impact on UVB irradiance





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Summary

- O_3 on a global scale will return to state from 1980 by 2050, i.e. faster than ODS, namely:
 - Around 2030-2040 in middle latitudes of both hemispheres
 - Around 2045-2060 in Antarctica
 - The increase of ozone will be accelerated in a colder stratosphere under the impact of GHGs
- As result, at the end of the 21st century the stratospheric ozone concentration will be higher than in 1980
- Tropical ozone might decrease due to climate-change induced changes in the Dobson-Brewer circulation
- Large uncertainties in future emission scenarios and resulting impacts

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