Mid-Latitude Stratospheric Chemistry

AOSC/CHEM 433 & AOSC/CHEM 633

Ross Salawitch

Class Web Sites:
http://www2.atmos.umd.edu/~rjs/class/spr2024
https://umd.instructure.com/courses/1358887

Today:

- **Background on CFCs**
- **Ozone Depletion Potenial**
- **Importance of how a chemical cycle is completed wrt odd-oxygen loss**
- **Role of halogens and aerosol loading on mid-latitude ozone**
- **Connection to recent research**

Lecture 15

2 April 2024

Motivation For Today

Total Column Ozone not recovering as fast as **"expected"**, particularly in NH mid-latitudes

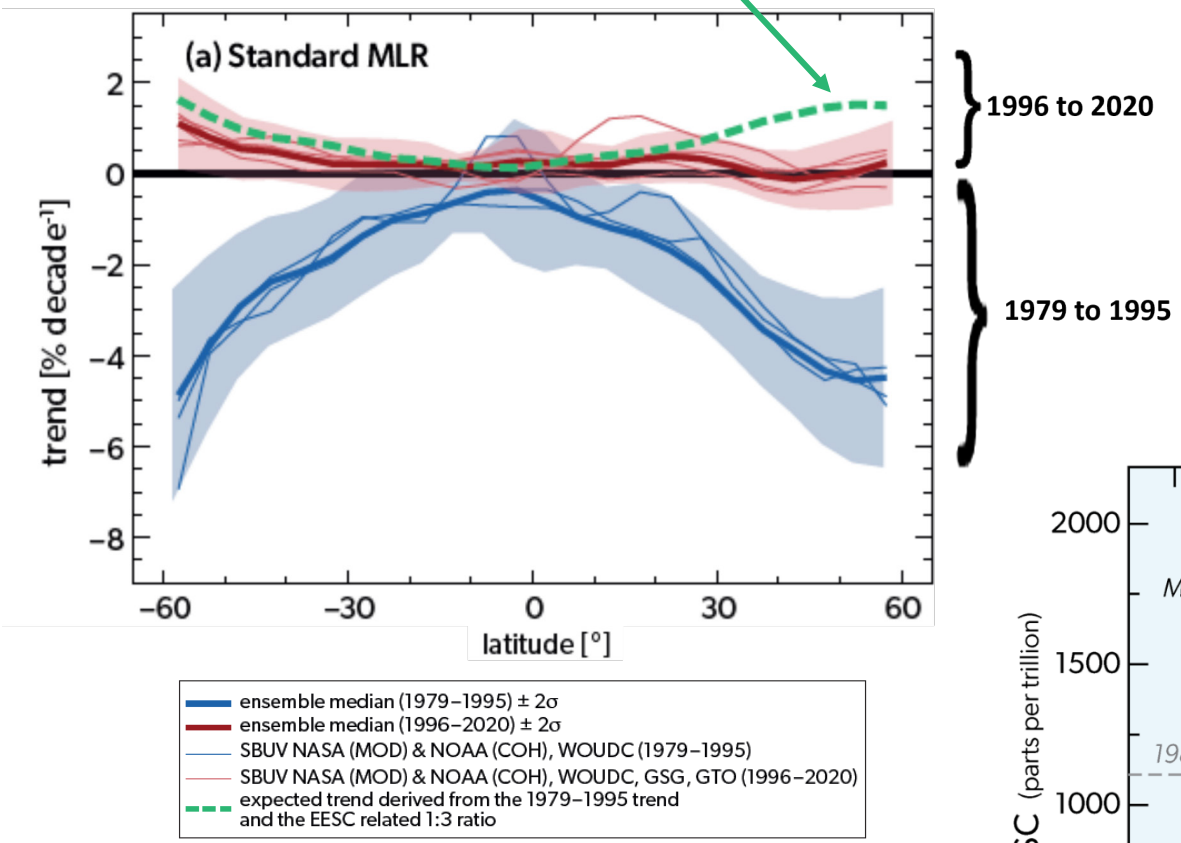
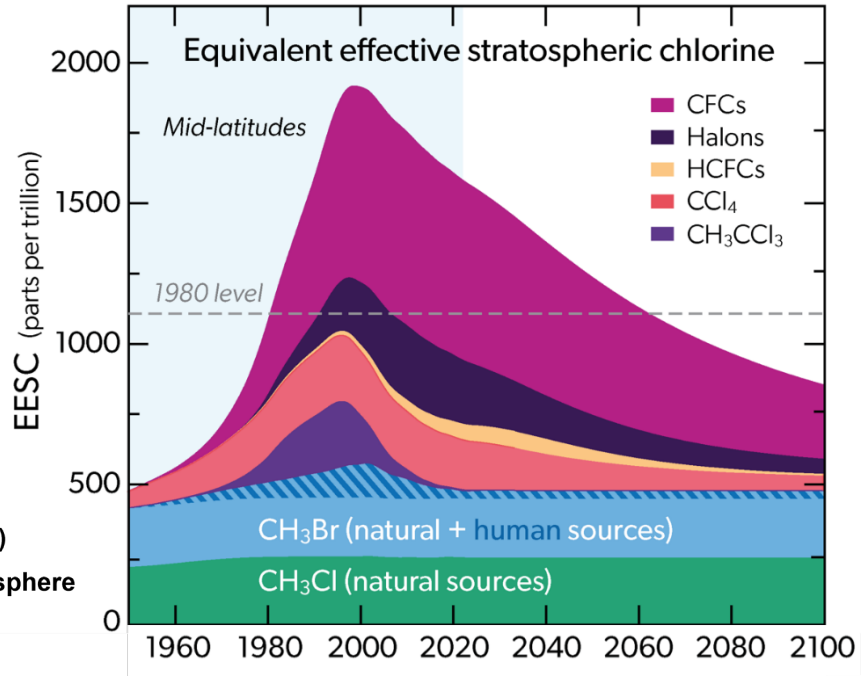


Fig 3-7, *WMO/UNEP Twenty QAs Ozone*



EESC = Cl_y + 60 × Br_y

where Cl_y and Br_y refer to inorganic (that is, reactive and reservoir) chlorine and bromine respectively in the midlatitude, lower stratosphere

Motivation For Today

Current Trajectory of Stratospheric Halogens *also* not recovering as fast as had once been **projected (Montreal 2007 line)**

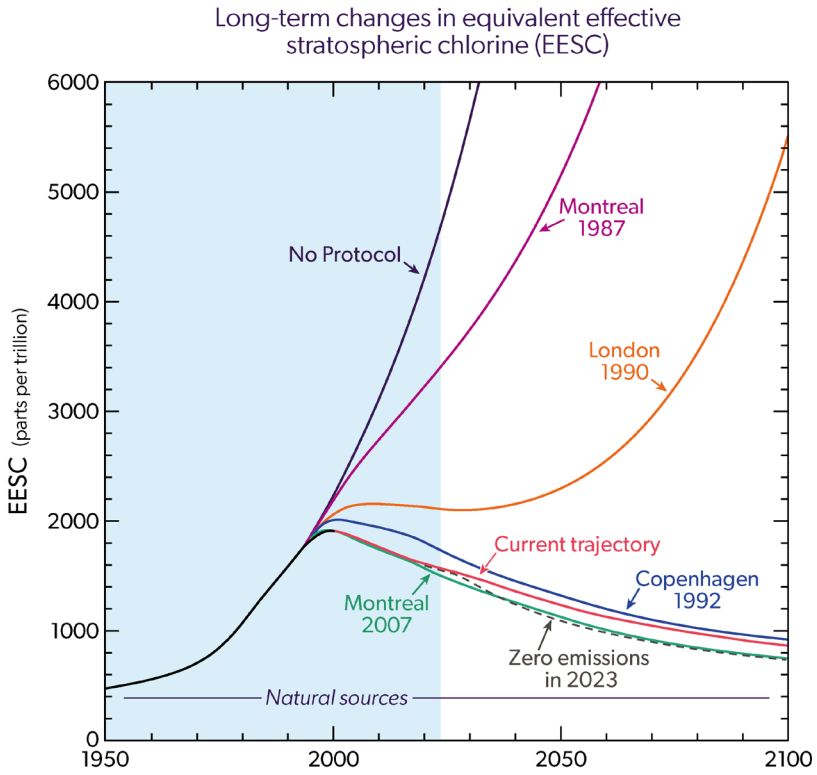
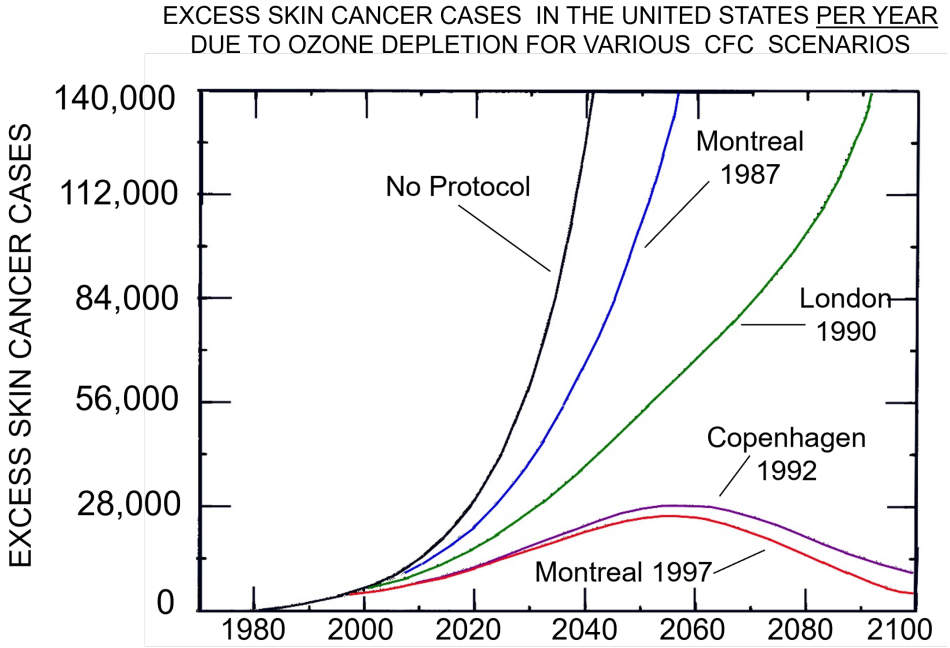
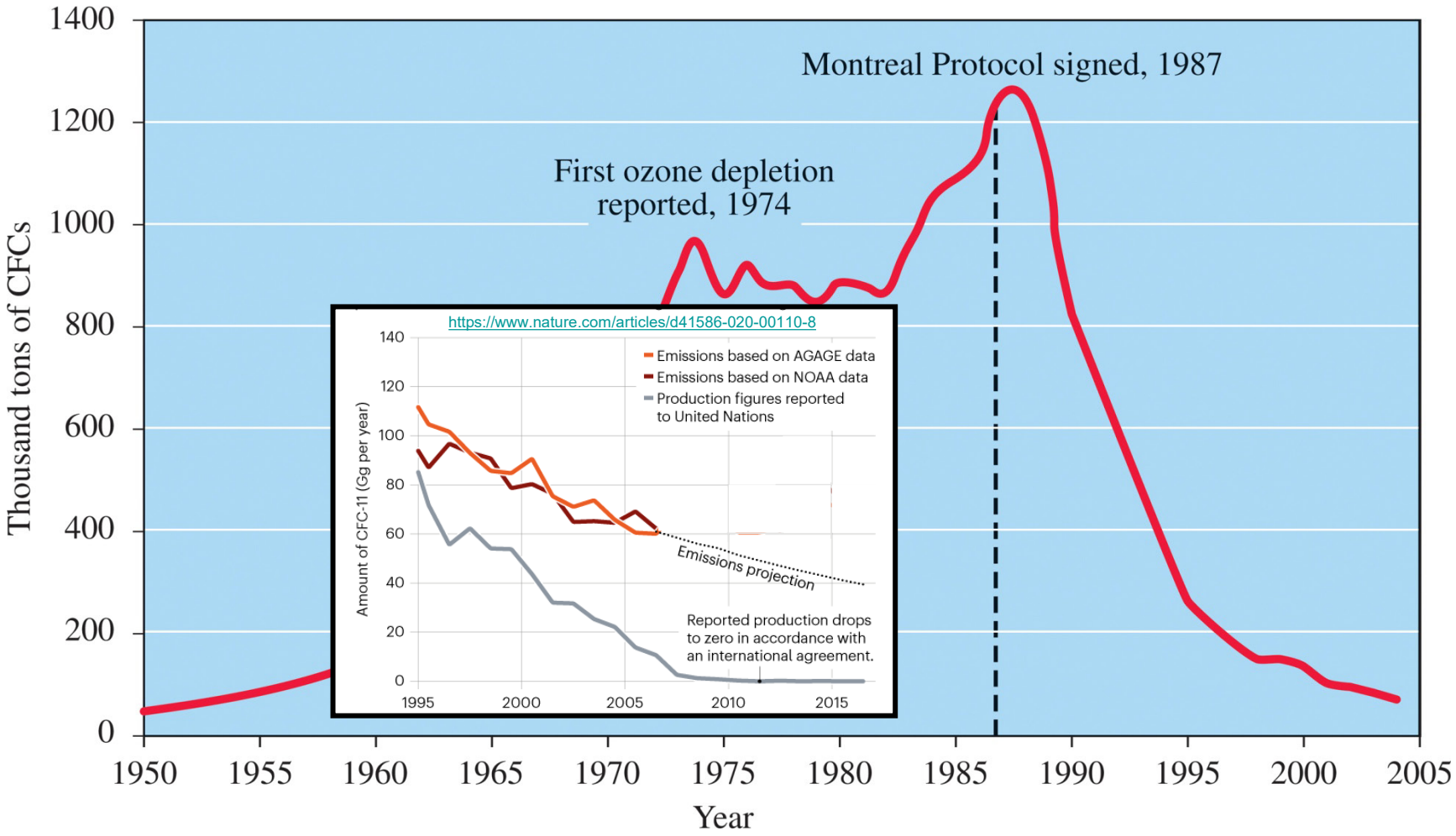


Fig Q14-1, *WMO/UNEP Twenty QAs Ozone*

Nonetheless, the Montreal Protocol has been a huge success and has prevented a runaway increase in skin cancer cases



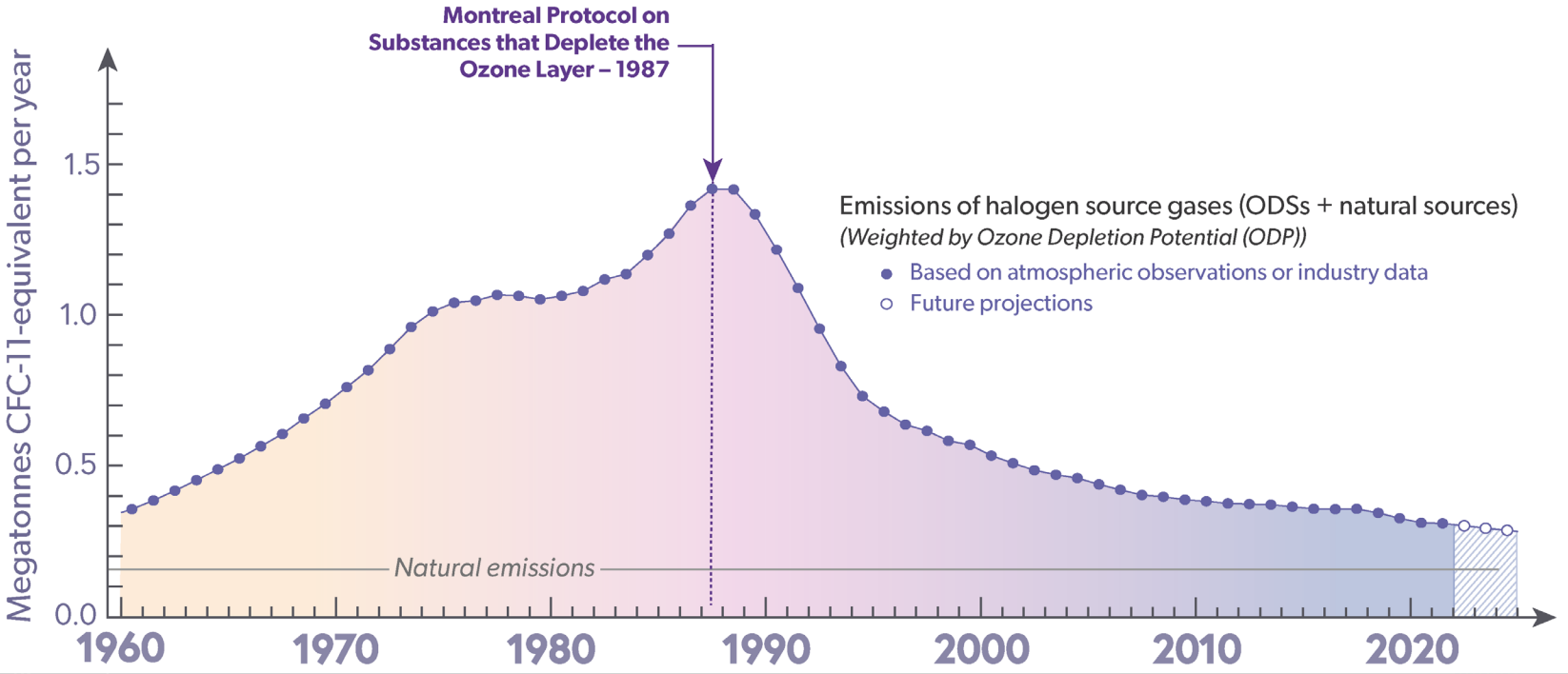
Montreal Protocol and Various Amendments Have Banned Industrial Production of CFCs and Halons



Global Production of CFCs, Fig. 2.19, Chemistry in Context

Montreal Protocol and Various Amendments Have Banned Industrial Production of CFCs and Halons

Global Emissions of all CFCs, **Fig Q0-1, *WMO/UNEP Twenty QAs Ozone***



Why was the introduction of Freon-12 as a refrigerant gas in the 1930s hailed as a great triumph?

What was the dire unintended consequence of the introduction of Freon-12?

Chlorine (left) & Bromine (right) Source Gases Entering the Stratosphere

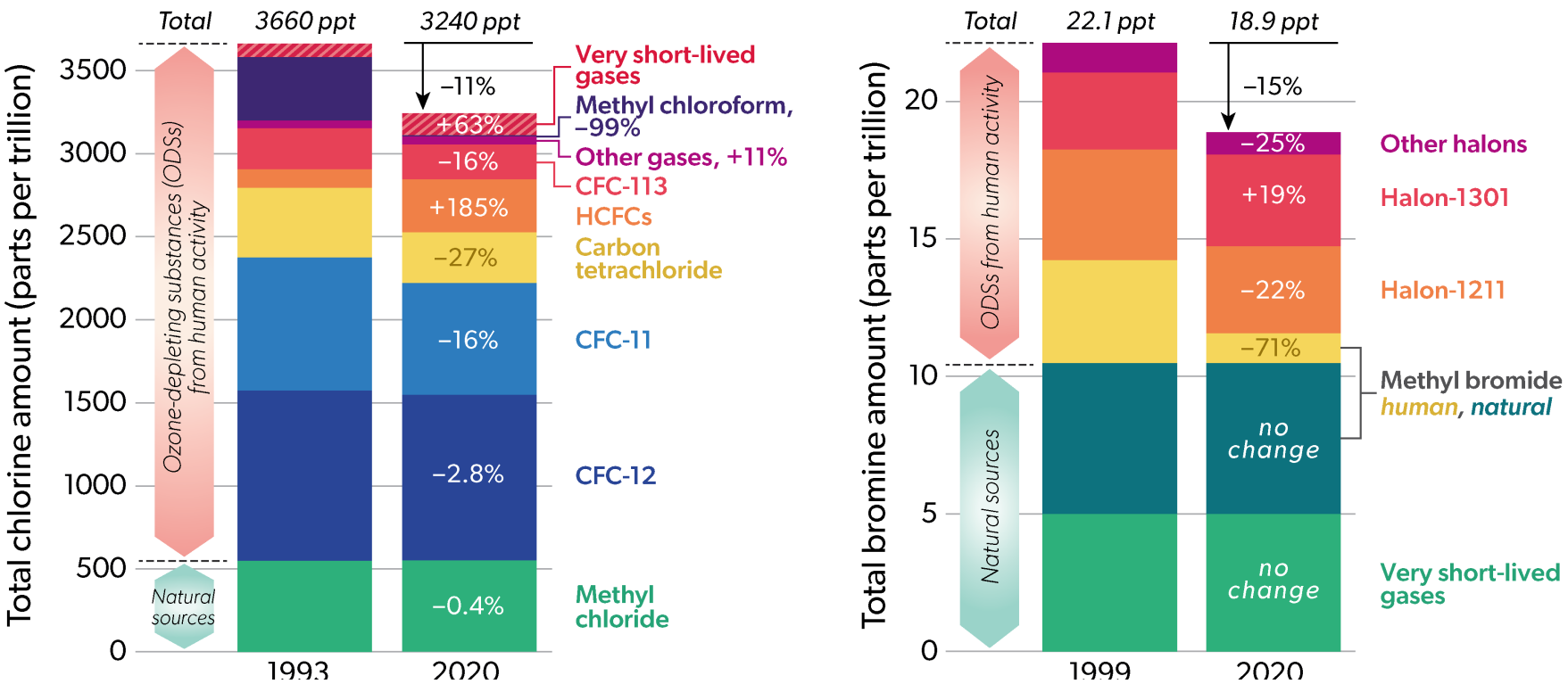


Fig Q6-1, *WMO/UNEP Twenty QAs Ozone*
https://csl.noaa.gov/assessments/ozone/2022/downloads/Chapter1_2022OzoneAssessment.pdf

Ozone Depletion Potential and Halocarbons

Table Q6-1. Atmospheric lifetimes, global emissions, Ozone Deletion Potentials, and Global Warming Potentials of some halogen source gases and HFC substitute gases.

Gas	Atmospheric Lifetime (years)	Ozone Depletion Potential (ODP)[b]
Halogen Source Gases		
Chlorine Gases		
CFC-11 (CCl_3F)	52	1
Carbon tetrachloride (CCl_4)	30	0.87
CFC-113 (CCl_2FCClF_2)	93	0.82
CFC-12 (CCl_2F_2)	102	0.75
Methyl chloroform (CH_3CCl_3)	5.0	0.12
HCFC-141b (CH_3CCl_2F)	8.8	0.102
HCFC-142b (CH_3CClF_2)	17	0.057
HCFC-22 (CHF_2Cl)	12	0.038
Methyl chloride (CH_3Cl)	0.9	0.015
Bromine Gases		
Halon-1301 ($CBrF_3$)	72	17
Halon-1211 ($CBrClF_2$)	16	7.1
Methyl bromide (CH_3Br)	0.8	0.57
Hydrofluorocarbons (HFCs)		
HFC-23 (CHF_3)	228	0
HFC-143a (CH_3CF_3)	52	0
HFC-125 (CHF_2CF_3)	31	0
HFC-134a (CH_2FCF_3)	14	0
HFC-32 (CH_2F_2)	5.3	0
HFC-152a (CH_3CHF_2)	1.5	0
HFO-1234yf (CF_3CFCH_2)	0.03	0

continuous

$$\text{ODP (species "}i\text{")} = \frac{\text{global loss of } O_3 \text{ due to unit mass emission of "}i\text{"}}{\text{global loss of } O_3 \text{ due to unit mass emission of CFC-11}}$$

continuous

$$\approx \frac{(\alpha\, n_{Br} + n_{Cl})}{3} \frac{\tau_i}{\tau_{\text{CFC-11}}} \frac{MW_{\text{CFC-11}}}{MW_i}$$

where :

τ is the global atmospheric lifetime

MW is the molecular weight

n is the number of chlorine or bromine atoms

α is the effectiveness of ozone loss by bromine relative to ozone loss by chlorine

Ozone Depletion

According to Section 2.8 of Chemistry in Context, how much depletion of stratospheric ozone at mid-latitudes (60°S to 60°N) has occurred?

According to the Question 12 of the WMO/UNEP QAs, how much depletion of the Global Total Ozone layer has occurred?

Also, state whether you are either "good" or "concerned" with the different estimates for depletion of the ozone layer given in Question 12 of the WMO/UNEP QAs, compared to Section 28 of Chemistry in Context (i.e, your answer to the prior question).

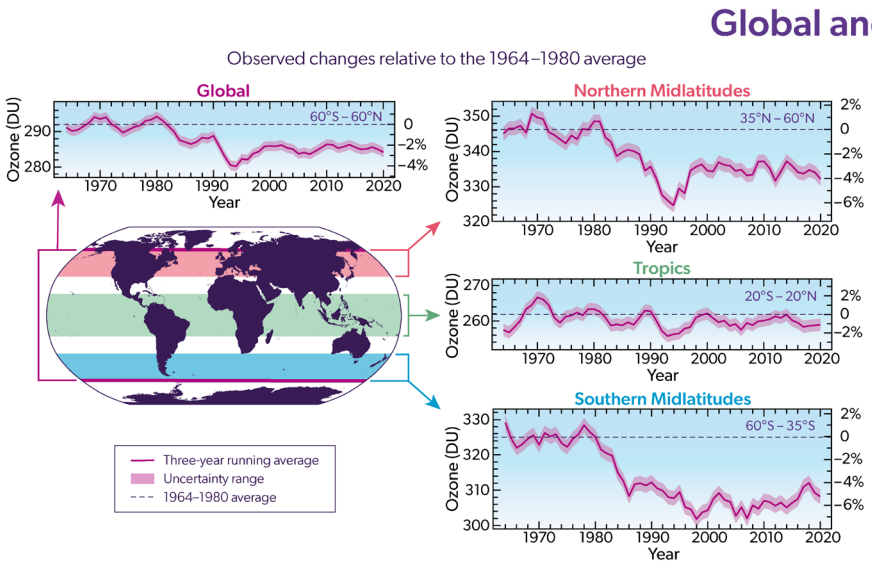


Fig Q12-1, *WMO/UNEP Twenty QAs Ozone*

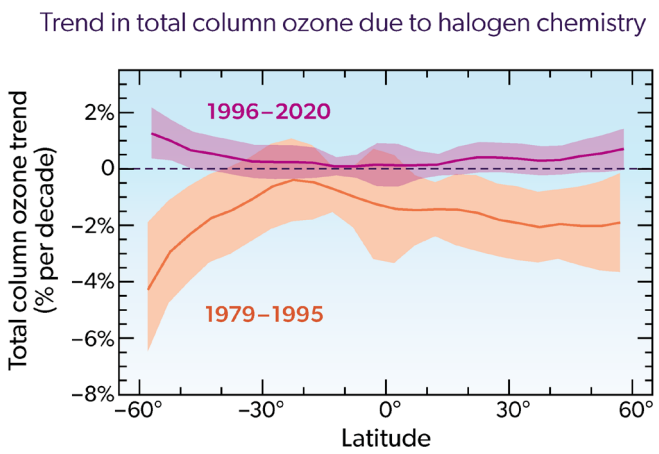


Fig Q12-2, *WMO/UNEP Twenty QAs Ozone*

Multiple Linear Regression Inputs and Outputs

Inputs

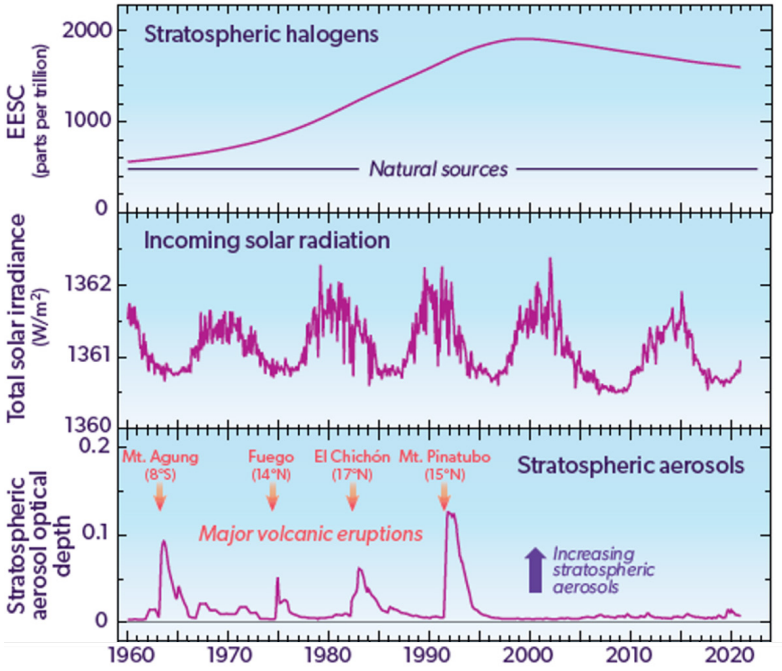
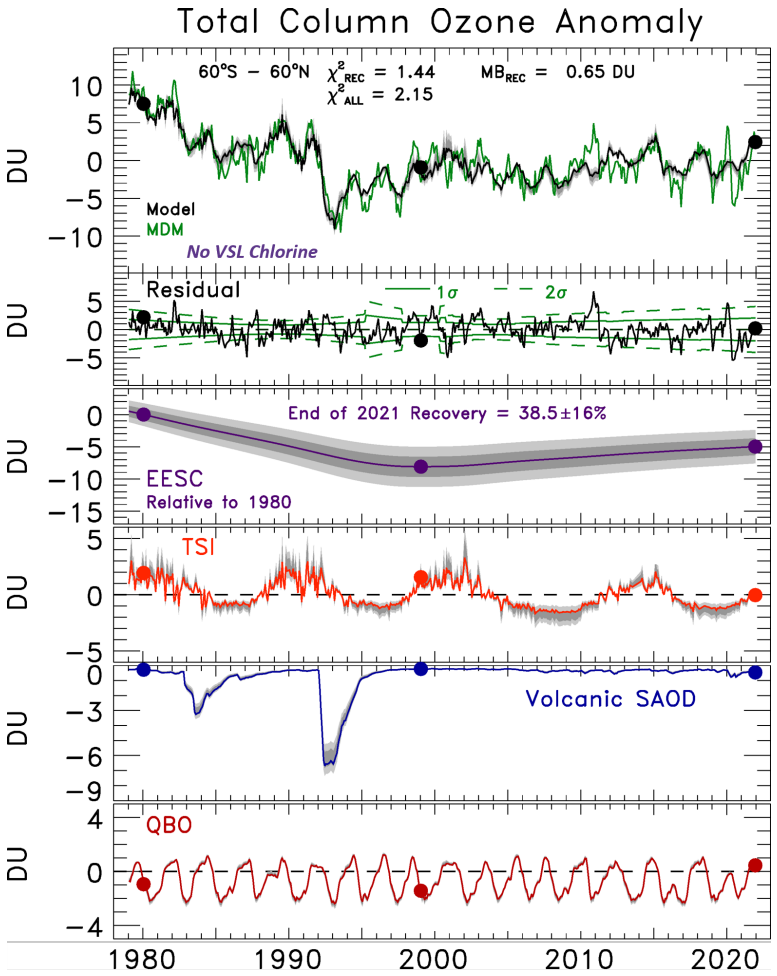


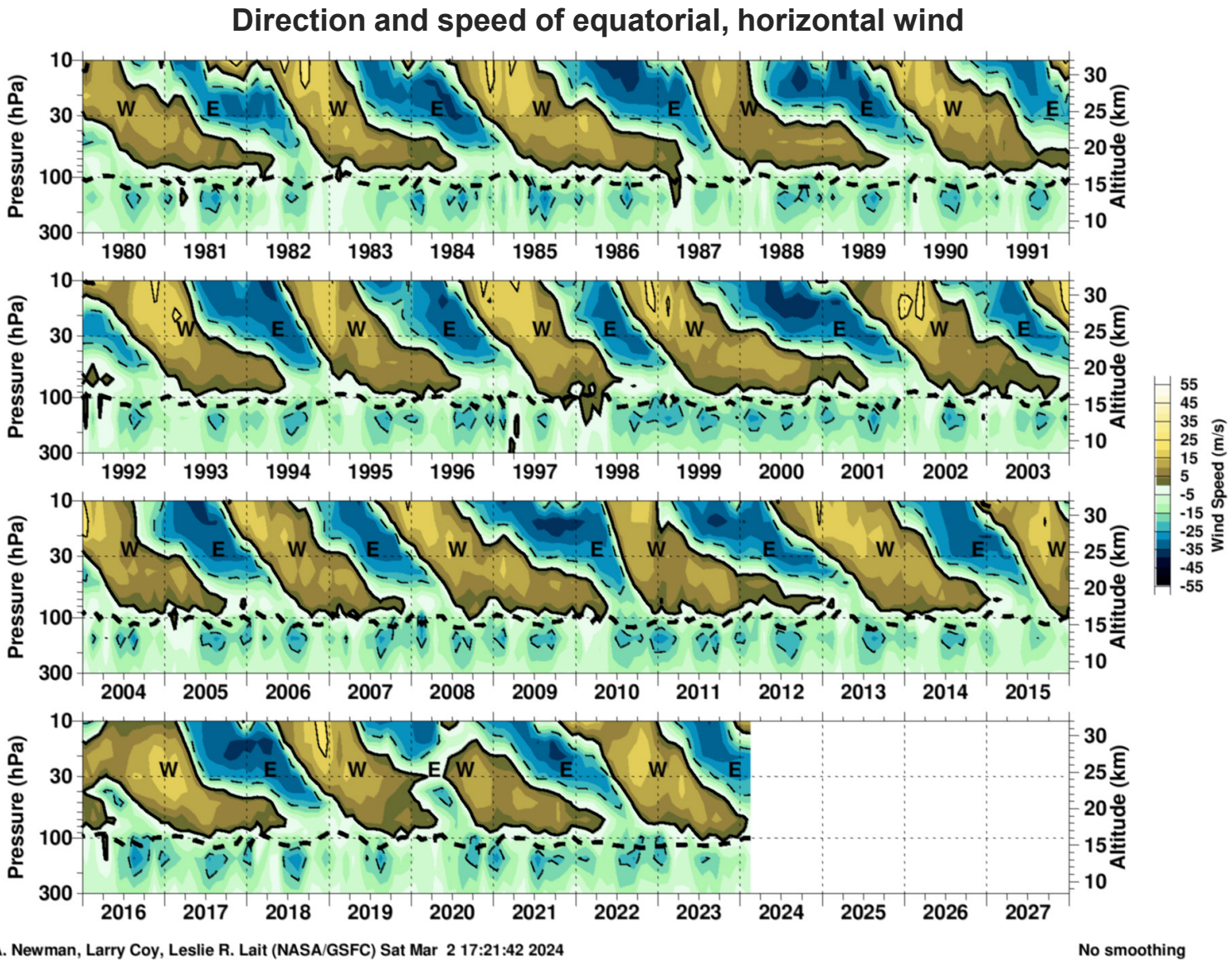
Fig Q13-1, *WMO/UNEP Twenty QAs Ozone*

Outputs



McBride *et al.*, Manuscript In Preparation, 2024

Quasi-Biennial Oscillation of Stratospheric Winds



https://acd-ext.gsfc.nasa.gov/Data_services/met/qbo/qbo.html

Chapman Chemistry

$$[O_3] = \left(\frac{J_1 \, k_2}{J_3 \, k_4} \right)^{1/2} f_{O2} \, [M]^{3/2}$$

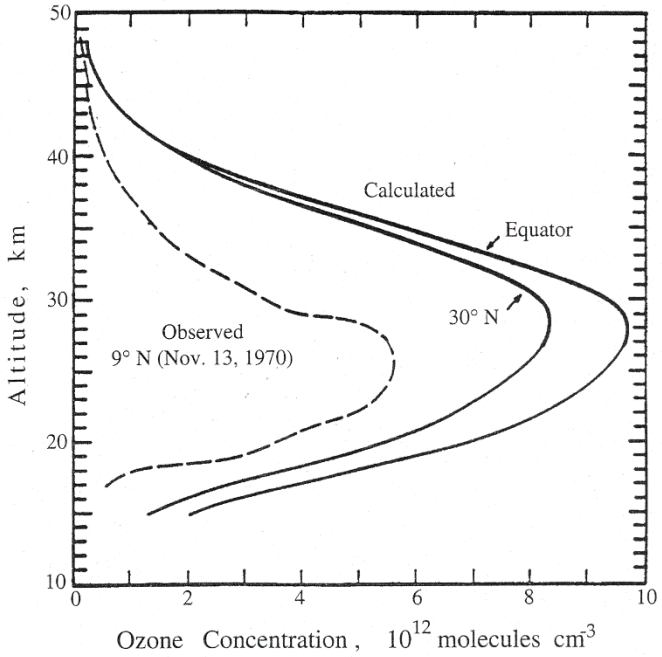


FIGURE 4.6 Comparison of stratospheric ozone concentrations as a function of altitude as predicted by the Chapman mechanism and as observed over Panama (9° N) on November 13, 1970.

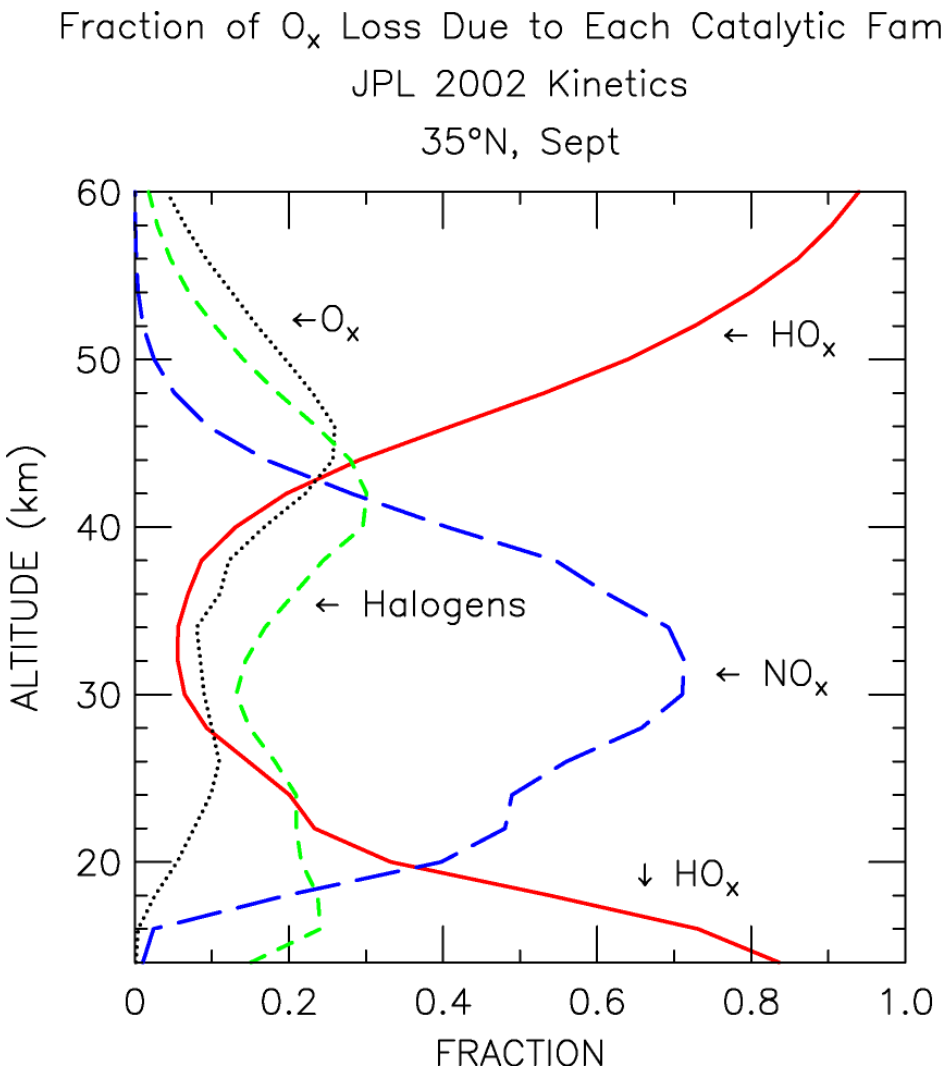
$[O_3]$ falls off with increasing altitude (high in stratosphere), at a rate determined by $[M]^{3/2}$, because:

$[O_3]$ falls off with decreasing altitude (low in stratosphere) due to a rapid drop in J_1, reflecting:

Observed $[O_3]$ < Chapman $[O_3]$: why ?!?

Lecture 10

Stratospheric Photochemistry: Odd Oxygen Loss By Families



Lecture 9

Calculated fraction of odd oxygen loss due to various families of radicals

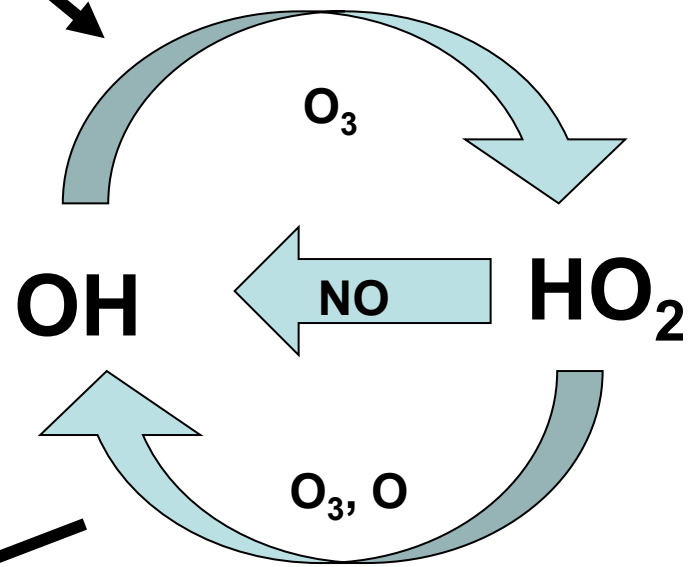
After Osterman *et al.*, *GRL*, 24, 1107, 1997;
Sen *et al.*, *JGR*, 103, 3571. 1998;
Sen *et al.*, *JGR*, 104, 26653, 1999.

HO_x : OH and HO_2

OH and HO_2 are central to stratospheric and tropospheric photochemistry

Production : $O^1D + H_2O \rightarrow OH + OH$

$O^1D + CH_4 \rightarrow OH + CH_3$



Loss: $OH + HO_2 \rightarrow H_2O + O_2$

$OH + HNO_3 \rightarrow H_2O + NO_3$

HO_x : OH and HO_2

OH and HO_2 are central to stratospheric and tropospheric photochemistry

Rapid inner cycle:

HO_2 formation:

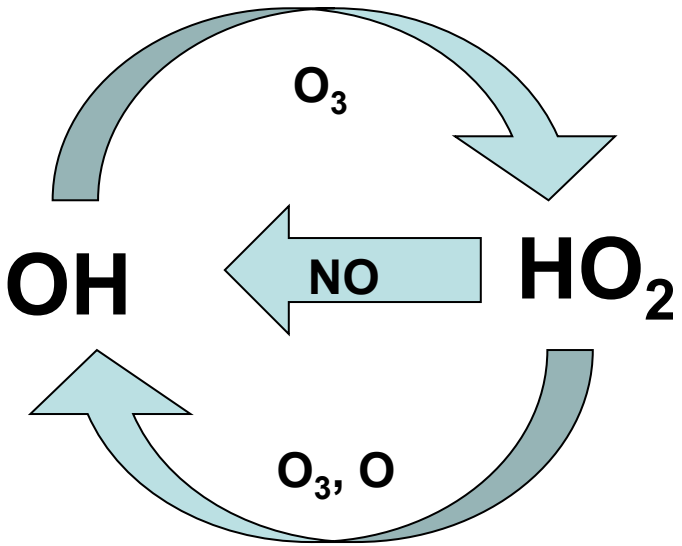
$$OH + O_3 \rightarrow HO_2 + O_2 \qquad (1)$$

HO_2 loss:

$$HO_2 + NO \rightarrow OH + NO_2 \qquad (2)$$

or $$HO_2 + O \rightarrow OH + O_2 \qquad (3)$$

or $$HO_2 + O_3 \rightarrow OH + O_2 + O_2 \qquad (4)$$



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HO_2 loss step (2):

$$\cancel{OH} + O_3 \rightarrow \cancel{HO_2} + O_2$$
$$\cancel{HO_2} + NO \rightarrow \cancel{OH} + NO_2$$

Net: $$O_3 + NO \rightarrow O_2 + NO_2$$

This is followed quickly by:

$$NO_2 + h\nu \rightarrow NO + O$$

Yielding final "net":

$$O_3 \rightarrow O + O_2$$

Null cycle

with respect to production & loss of odd oxygen

HO_x : OH and HO_2

OH and HO_2 are central to stratospheric and tropospheric photochemistry

Rapid inner cycle:

HO_2 formation:

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HO_2 loss step (3):

$$OH + O_3 \rightarrow HO_2 + O_2$$
$$HO_2 + O \rightarrow OH + O_2$$
$$\text{Net: } O_3 + O \rightarrow O_2 + O_2$$

HO_2 loss step (4):

$$OH + O_3 \rightarrow HO_2 + O_2$$
$$HO_2 + O_3 \rightarrow OH + O_2 + O_2$$
$$\text{Net: } O_3 + O_3 \rightarrow O_2 + O_2 + O_2$$

Catalytic Ozone (Odd Oxygen) Loss Cycles

Odd Oxygen Loss - HO_x

$$\frac{d\,(\text{Odd Oxygen})}{dt} = -2\,k_4\,[HO_2][O_3] - 2\,k_3\,[HO_2][O] \qquad \text{Eq (7)}$$

The reactions:

$$HO_2 + O \rightarrow OH + O_2 \qquad (3)$$
$$HO_2 + O_3 \rightarrow OH + O_2 + O_2 \qquad (4)$$

are rate limiting steps for O_3 loss by two catalytic cycles:

Cycle (1) Net :

$$O_3 + O \rightarrow 2\,O_2$$

Cycle (2) Net :

$$O_3 + O_3 \rightarrow 3\,O_2$$

As a convenient short hand, **we consider HO_2 to be odd oxygen**

Then:

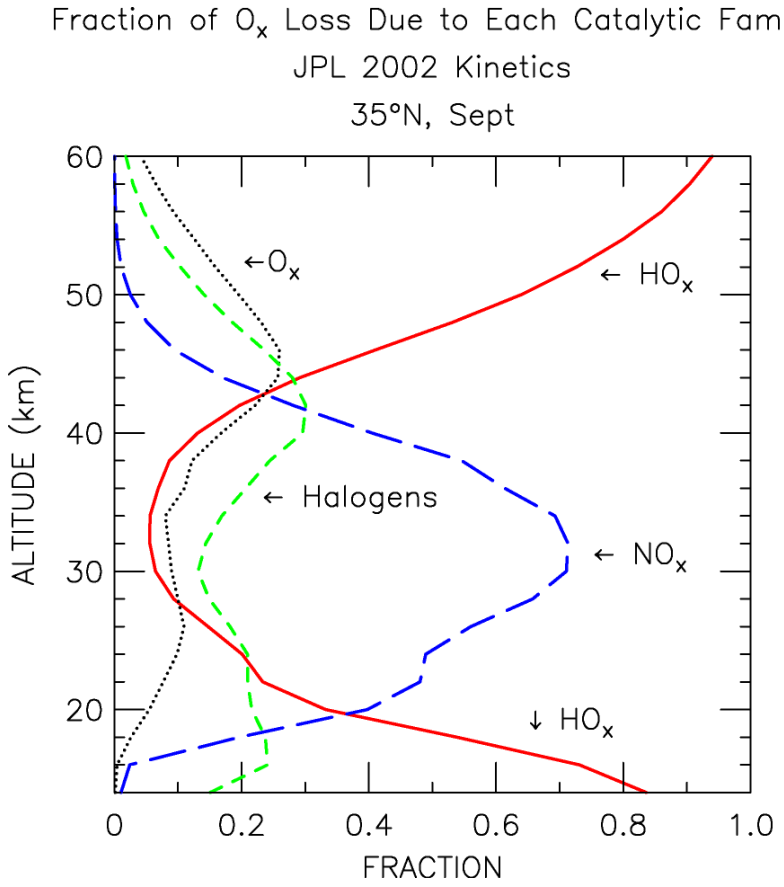
clear now that reactions (3) and (4) each consume two odd oxygens at rates determined by $2\,k_3\,[HO_2]\,[O]$ and $2\,k_4\,[HO_2][O_3]$

Odd Oxygen Loss - HO_x

At what altitudes will loss of ozone by these rate limiting steps be dominant ?

$$HO_2 + O \rightarrow OH + O_2 \qquad (3)$$
$$HO_2 + O_3 \rightarrow OH + O_2 + O_2 \qquad (4)$$

One dominates at low altitude, the other at high altitude ⇒ which is which ?!?



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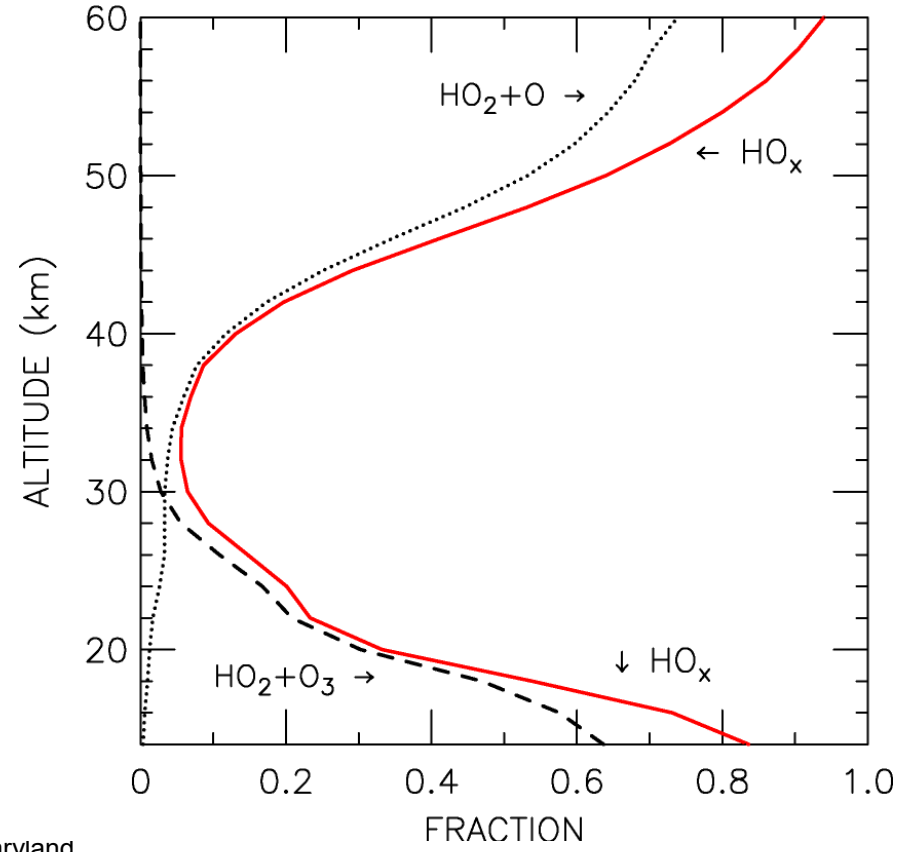
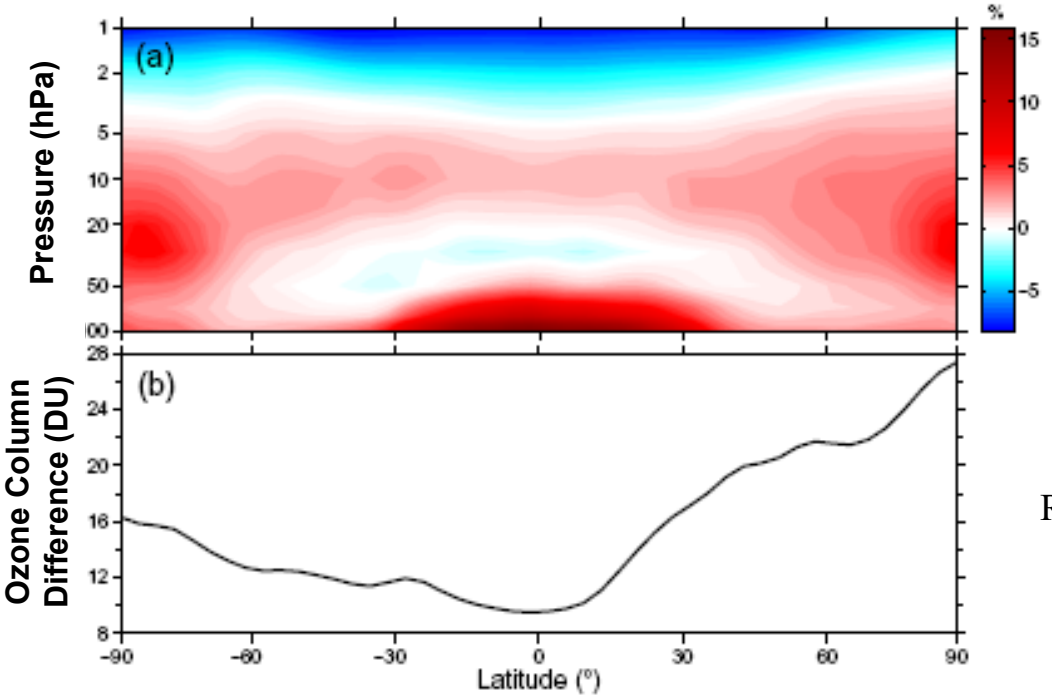


CH_4 and Stratospheric Ozone



Revell *et al.*, *ACP*, 2012

Stratospheric O_3 difference in the 2090s found for a computer simulation run using CH_4 from RCP 8.5 minus that of a simulation using CH_4 from RCP 2.6

Rising CH_4 leads to:

a) ozone loss in the upper stratosphere by increasing the speed of OH and HO_2 (HO_x) mediated loss cycles.

b) a cooler stratosphere, slowing the rate of all ozone loss cycles

c) speeds up the rate of Cl+CH_4, shifting chlorine from ClO into HCl (i.e., deactivates chlorine)

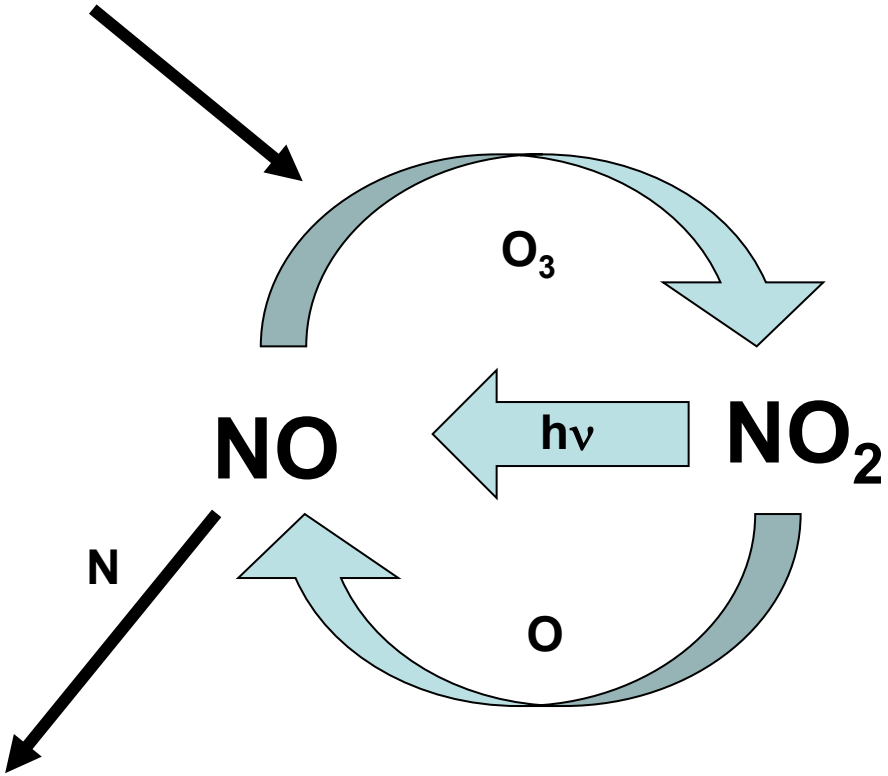
d) more HO_2 in the lowermost stratosphere where there is sufficient CO to result in O_3 production by "smog chemistry"

Computer models project stratospheric column O_3 will increase as CH_4 rises

NO_x : NO and NO_2

NO and NO_2 are central to stratospheric and tropospheric photochemistry

Stratospheric **Production : $O^1D + N_2O \rightarrow NO + NO$**



Final sinks : $N + NO \rightarrow N_2 + O$ (uppermost stratosphere)
HNO_3 solubility & rainout (lowermost stratosphere)

NO_x : NO and NO_2

NO and NO_2 are central to stratospheric and tropospheric photochemistry

Rapid inner cycle:

NO_2 formation:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (1)$$

NO_2 loss:

$$NO_2 + h\nu \rightarrow NO + O \qquad (2)$$

or $$NO_2 + O \rightarrow NO + O_2 \qquad (3)$$

NO_2 loss step (2):

$$\begin{aligned} NO + O_3 &\rightarrow NO_2 + O_2 \\ NO_2 + h\nu &\rightarrow NO + O \\ \hline \text{Net:}\quad O_3 + h\nu &\rightarrow O + O_2 \end{aligned}$$

NO_2 loss step (3):

$$\begin{aligned} NO + O_3 &\rightarrow NO_2 + O_2 \\ NO_2 + O &\rightarrow NO + O_2 \\ \hline \text{Net:}\quad O_3 + O &\rightarrow 2\,O_2 \end{aligned}$$

Can show:

$$\frac{dO_3}{dt} + \frac{dO}{dt} = \frac{d\,(\text{Odd Oxygen})}{dt} = -2\,k_3\,[NO_2][O]$$

As a convenient short hand, **we consider NO_2 to be odd oxygen**

N_2O and NO_y

Loss of N_2O occurs mainly in the stratosphere due to:

photolysis – main sink

reaction with electronically excited $O(^1D)$ – minor sink

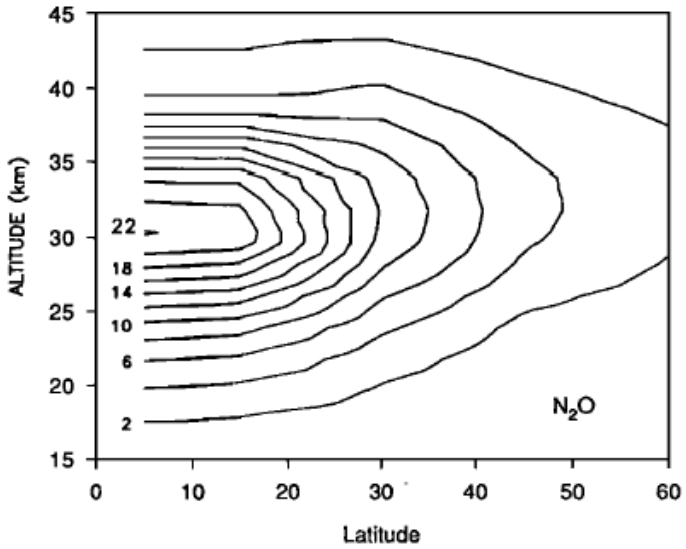


Fig. 11. Diurnally averaged loss rate for N_2O (10^2 molecules cm^{-3} s^{-1}) as a function of altitude and latitude, calculated with the line-by-line model, for equinox. The loss rate includes destruction of N_2O by reaction with $O(^1D)$ as well as photolysis.

Minschwaner, Salawitch, and McElroy, *JGR*, 1993

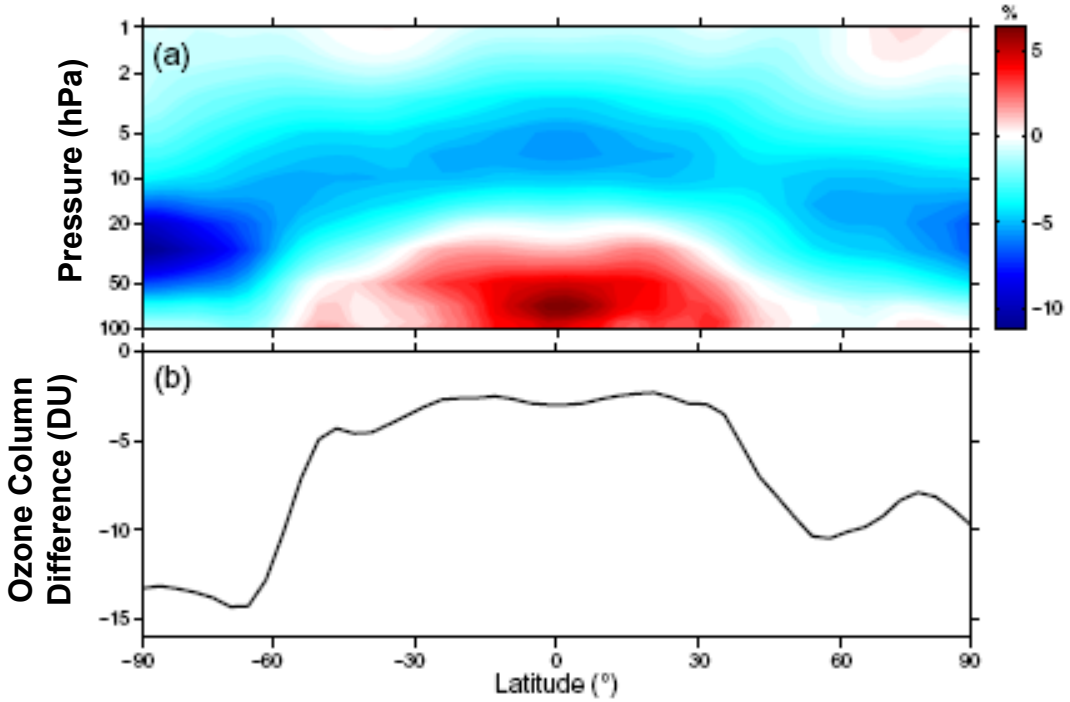
The minor sink for N_2O loss has a path that results in "reactive nitrogen": **Lecture 6**

$N_2O + O(^1D) \rightarrow NO + NO$

Reactive nitrogen (NO_y) is crucial to stratospheric chemistry

Oxides of nitrogen catalyze loss of stratospheric O_3 & participate in a series of chemical reactions that affect partitioning of hydrogen and chlorine radicals, etc.

N_2O and Stratospheric Ozone



Revell *et al.*, *ACP*, 2012

Stratospheric O_3 difference in the 2090s found for a computer simulation run using N_2O from RCP 8.5 minus that of a simulation using N_2O from RCP 2.6

Rising N_2O leads to:

a) ozone loss in the middle & upper stratosphere by increasing the speed of NO and NO_2 (NO_x) mediated loss cycles.

b) speeds up the rate of $OH+NO_2+M \rightarrow HNO_3+M$ & $ClO+NO_2+M \rightarrow ClNO_3+M$ in the lowermost stratosphere, leading to slower ozone loss by these cycles & therefore more O_3 where these cycles dominate total loss of O_3

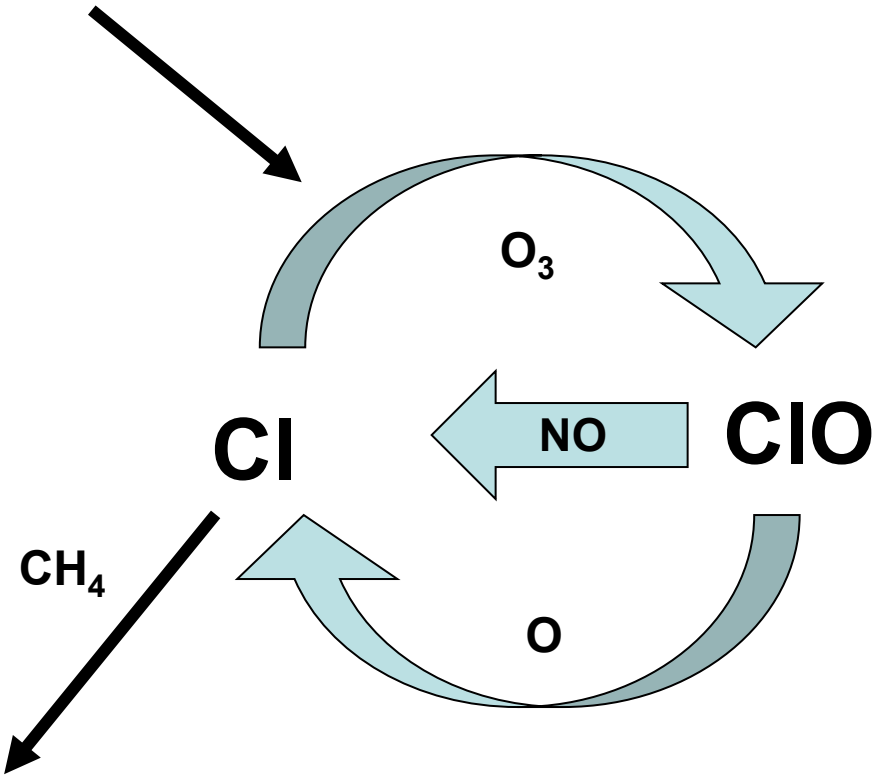
Computer models project stratospheric column O_3 will decline as N_2O rises

Lecture 6

ClO_x : ClO and Cl

ClO is central to stratospheric photochemistry, at mid-latitudes and polar regions

Production : CFCs +hν→ Inorganic chlorine



Final sinks : HCl solubility & rainout (lowermost stratosphere)

ClO_x : ClO and Cl

ClO is central to <u>stratospheric</u> photochemistry, at mid-latitudes and polar regions:

Rapid inner cycle:

ClO formation:

$$Cl + O_3 \rightarrow ClO + O_2 \qquad (1)$$

ClO loss:

$$ClO + NO \rightarrow Cl + NO_2 \qquad (2)$$

or $$ClO + O \rightarrow Cl + O_2 \qquad (3)$$

ClO loss step (2):

$$Cl + O_3 \rightarrow ClO + O_2$$
$$ClO + NO \rightarrow Cl + NO_2$$

Net: $O_3 + NO \rightarrow NO_2 + O_2$

Followed by: $NO_2 + h\nu \rightarrow NO + O$

Final net: $O_3 + h\nu \rightarrow O + O_2$

ClO loss step (3):

$$Cl + O_3 \rightarrow ClO + O_2$$
$$ClO + O \rightarrow Cl + O_2$$

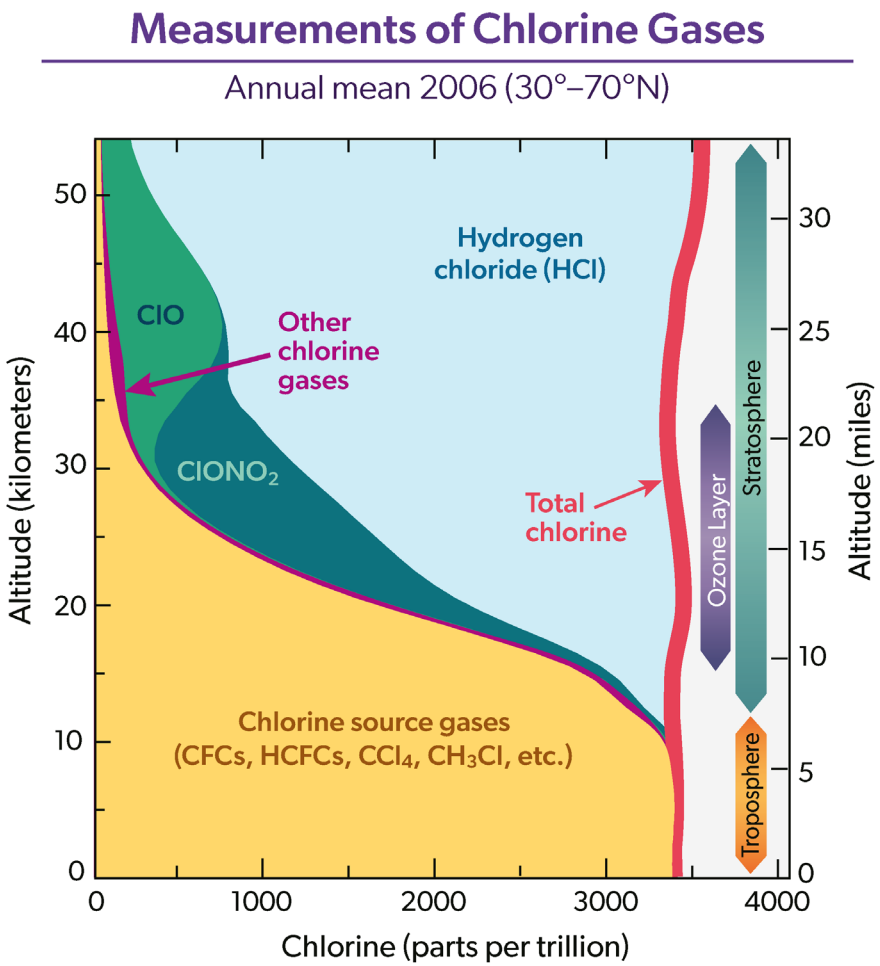
Net: $O_3 + O \rightarrow 2\,O_2$

Can show:

$$\frac{dO_3}{dt} + \frac{dO}{dt} = \frac{d\,(\text{Odd Oxygen})}{dt} = -2\,k_3\,[ClO][O]$$

As a convenient short hand, **we consider ClO to be odd oxygen**

Proof Halocarbons Reach The Stratosphere



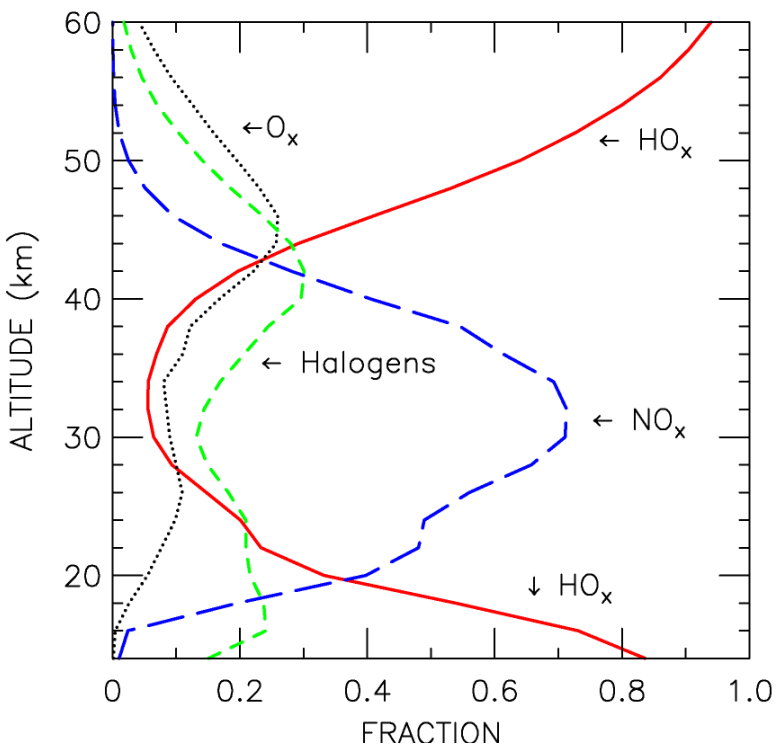
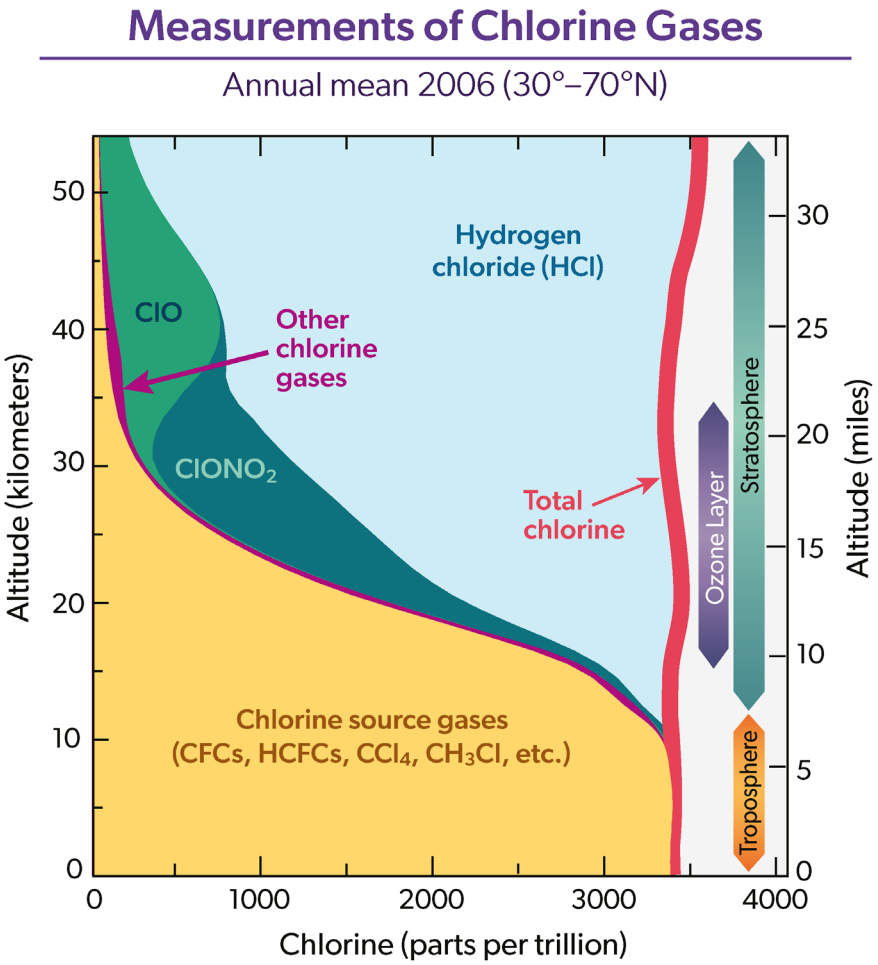


Fig Q7-2, *WMO/UNEP Twenty QAs Ozone*

Proof Halocarbons Reach The Stratosphere



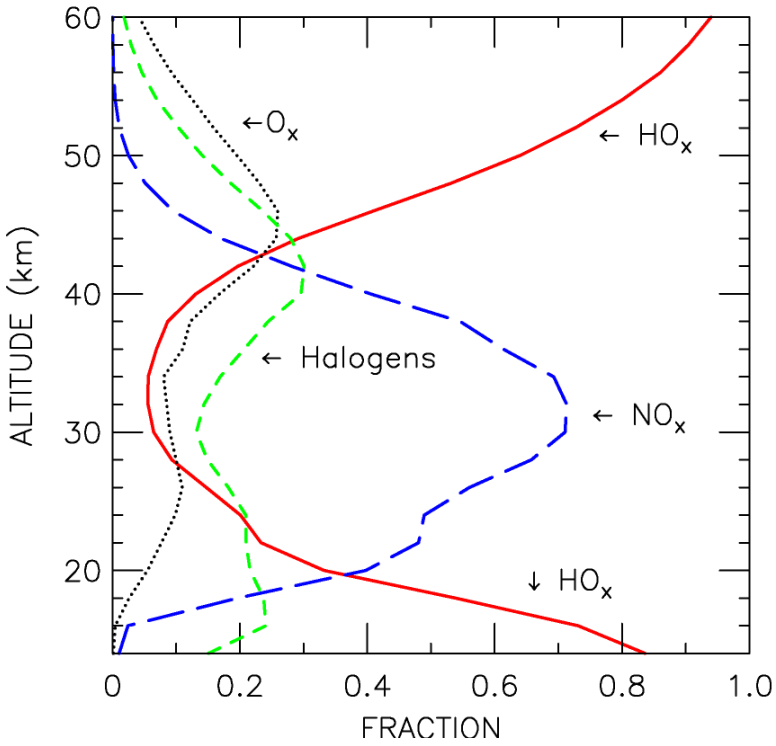


Fig Q7-2, *WMO/UNEP Twenty QAs Ozone*

Trends in Ozone, ~40 km

Line: range of model calculations,
where models are forced by
changing levels of stratospheric halogens

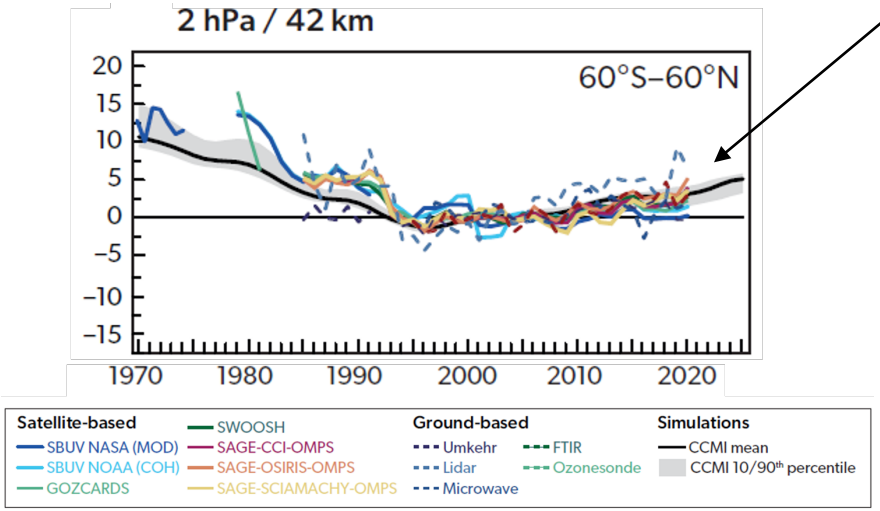
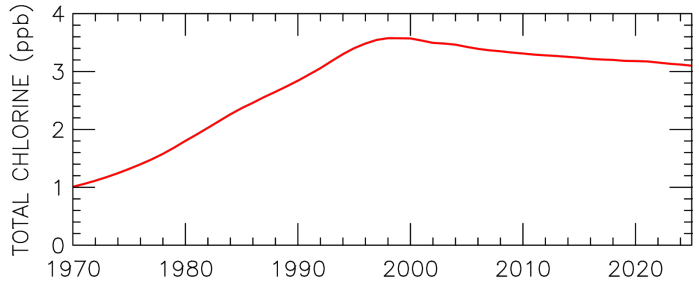
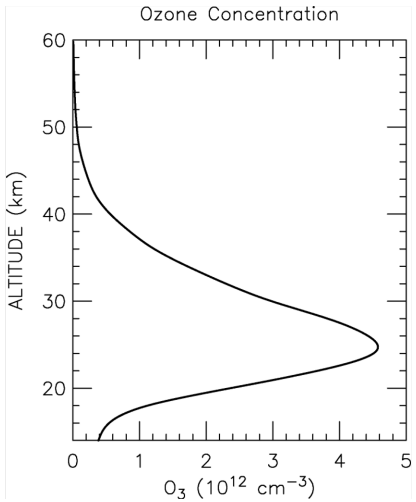


Fig 3-9, *WMO/UNEP Ozone Report*

Trends in ozone at 40 km are "well understood":
ozone is *anti-correlated* with time history
of upper stratospheric chlorine loading

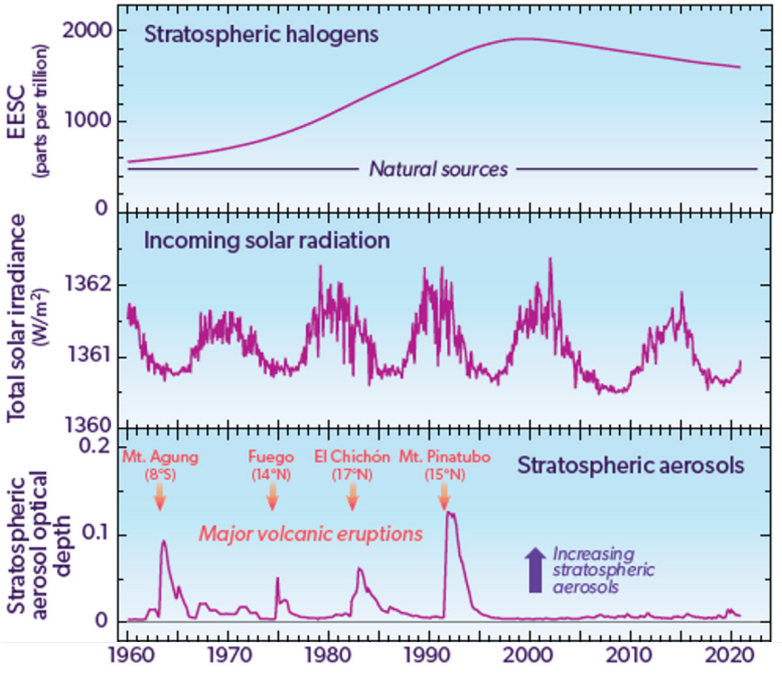


But: ozone at 40 km has little effect on surface UV radiation



Multiple Linear Regression Inputs and Outputs

Inputs



Outputs

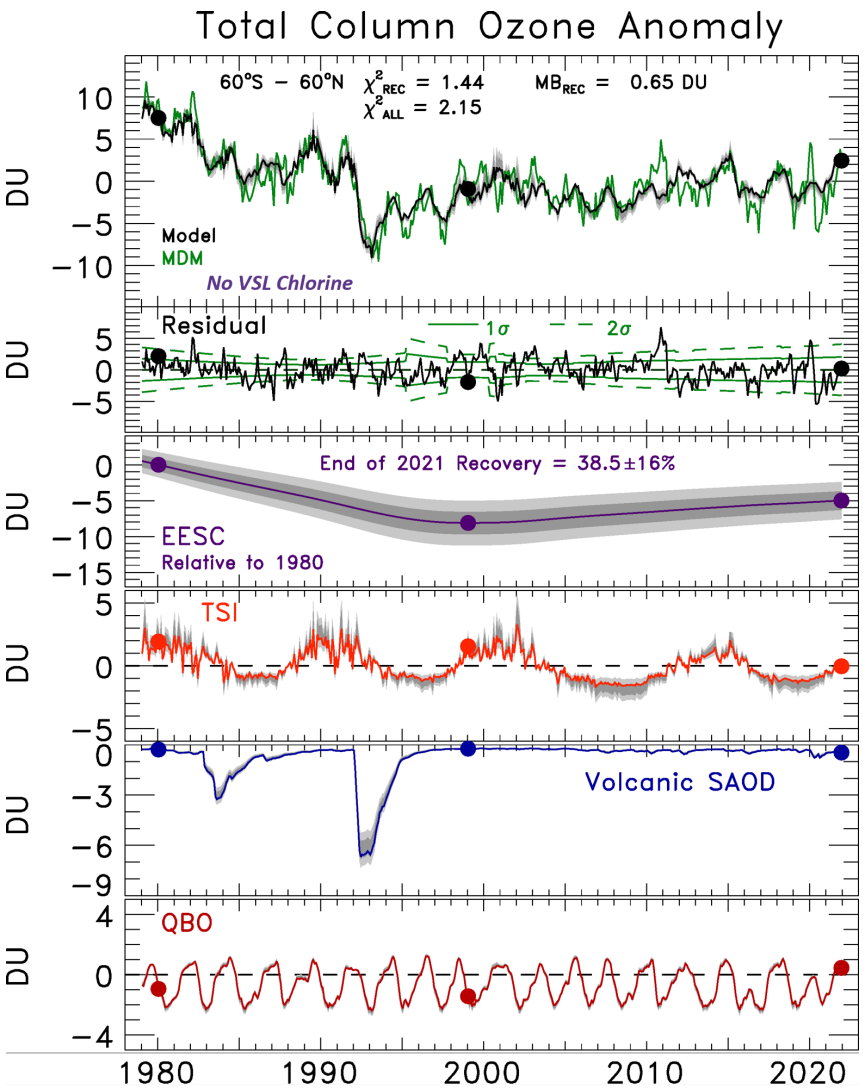
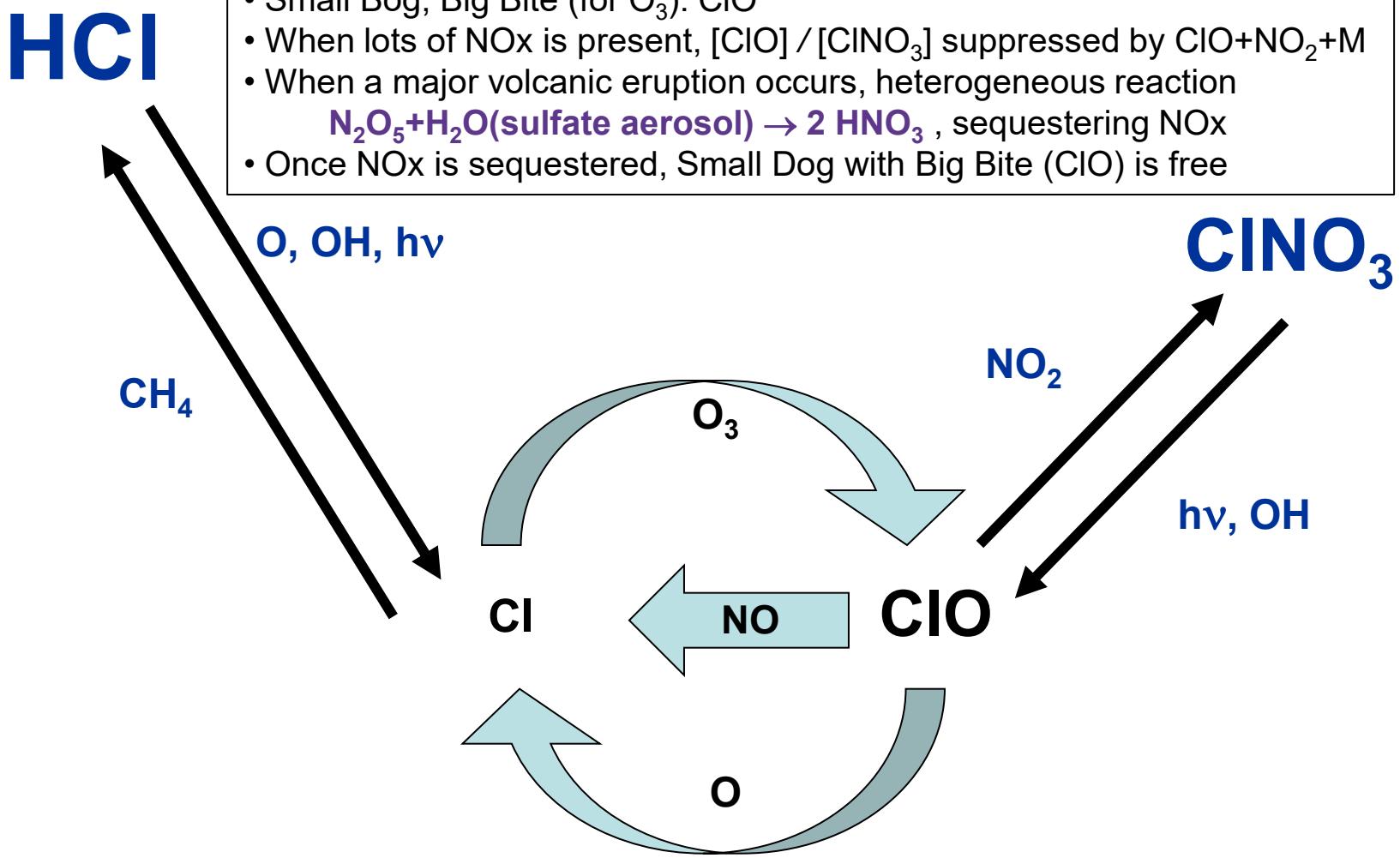


Fig Q13-1, *WMO/UNEP Twenty QAs Ozone*

Chemical reaction on surface of volcanic aerosol couples NO_2 and HNO_3

- As sulfate aerosol rises, NO_x (NO and NO_2) falls
- As NO_2 drops, $ClNO_3$ falls and ClO rises

- Big Dogs in Chlorine Family: HCl & $ClNO_3$
- Small Bog, Big Bite (for O_3): ClO
- When lots of NOx is present, [ClO] / [$ClNO_3$] suppressed by ClO+NO_2+M
- When a major volcanic eruption occurs, heterogeneous reaction
  $N_2O_5+H_2O(\text{sulfate aerosol}) \rightarrow 2\ HNO_3$, sequestering NOx
- Once NOx is sequestered, Small Dog with Big Bite (ClO) is free



Response of Stratospheric Constituents to Mount Pinatubo: aka The Rise (ClO) and Fall (NO_2) of Radicals

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 99, NO. D9, PAGES 18,861–18,869, SEPTEMBER 20, 1994

A two-dimensional modeling study of the volcanic eruption of Mount Pinatubo

S. Bekki and J. A. Pyle
Centre for Atmospheric Science, Department of Chemistry, University of Cambridge, Cambridge, England

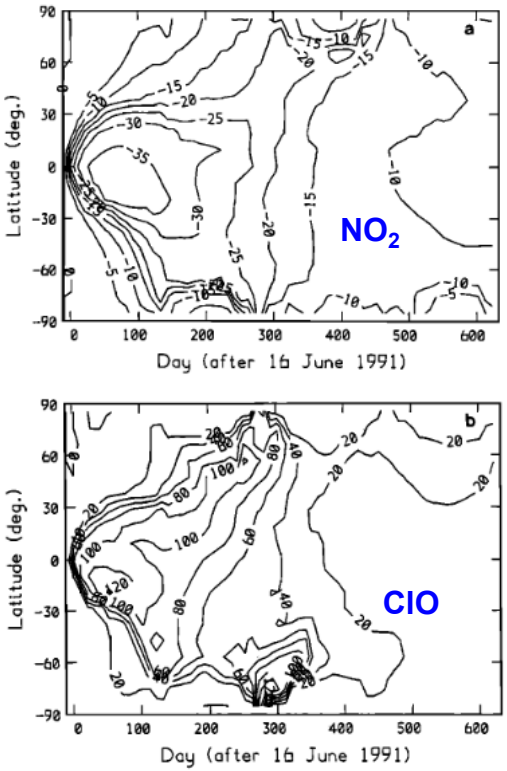


Figure 6. Percentage changes in (a) NO_2 and (b) ClO columns as a function of time and latitude for the volcanic run relative to the background run.

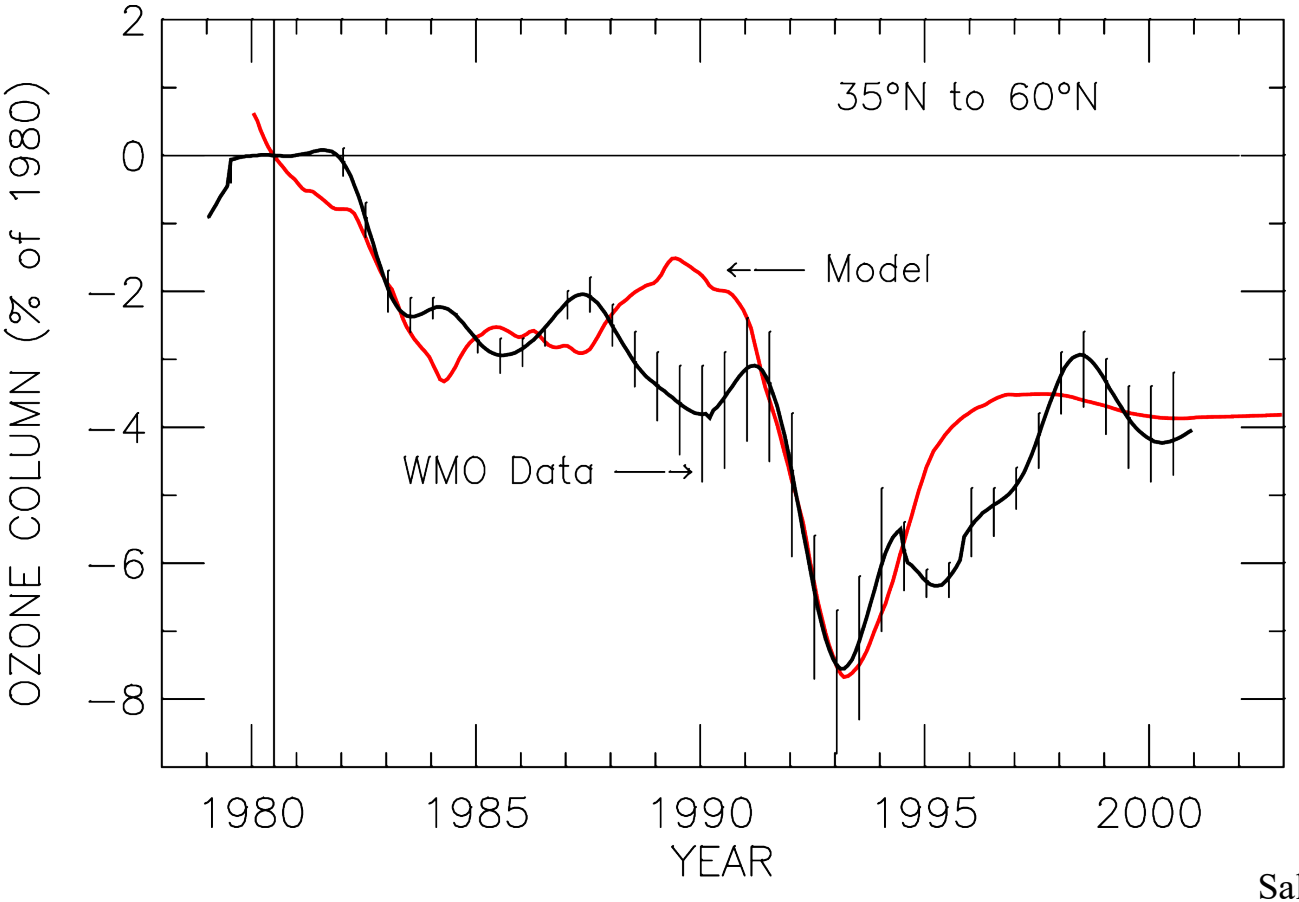
Abundance of stratospheric ozone responds to:

a) rise and fall of chlorine

b) volcanic perturbations to aerosol loading

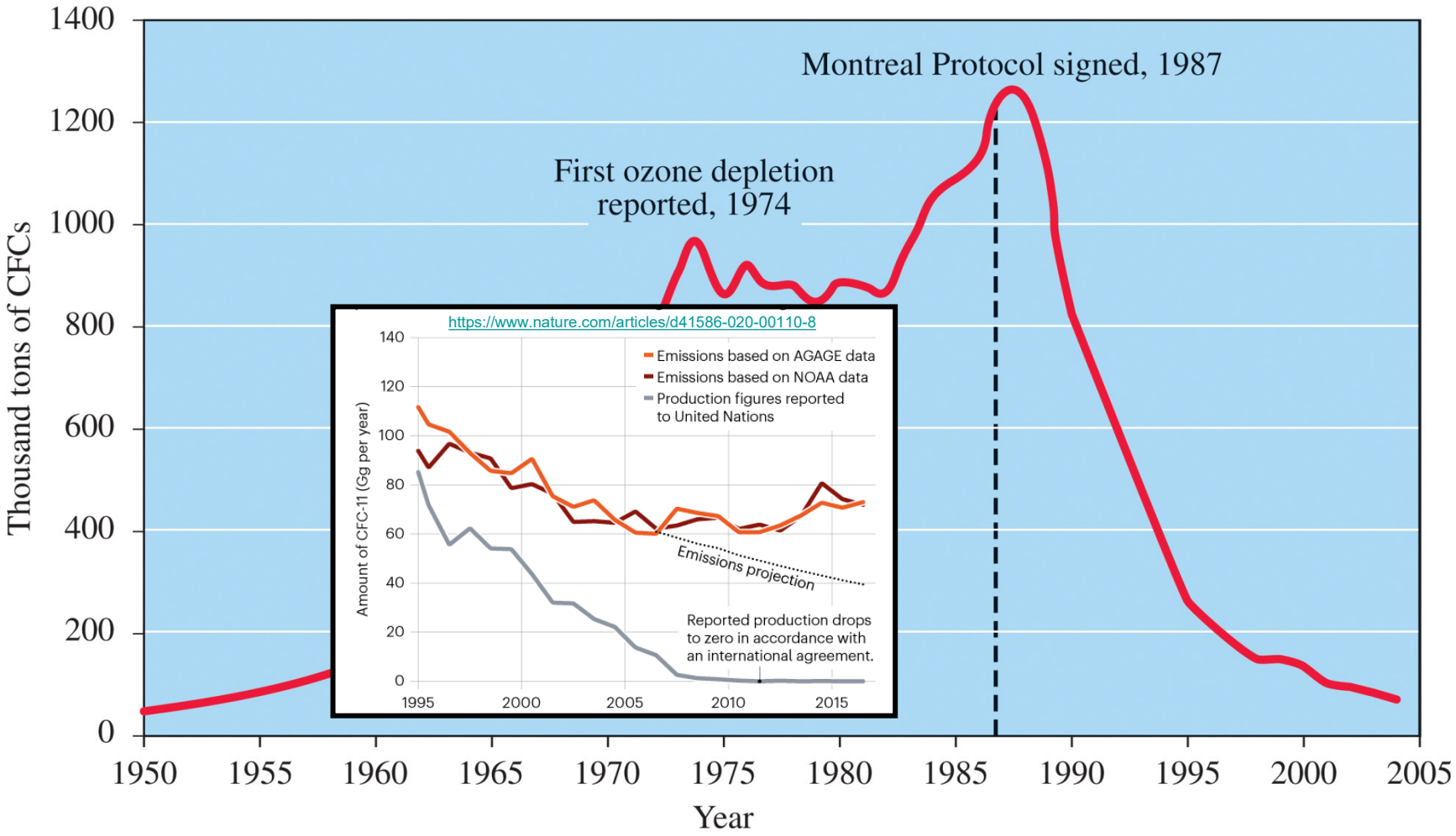
c) measured & modeled ozone column anomaly (deviation from mean)

simulated quite after June 1991 eruption of Mount Pinatubo, particularly in NH



Salawitch *et al.*, *GRL*, 2005

Montreal Protocol and Various Amendments Have Banned Industrial Production of CFCs and Halons



Global Production of CFCs, Fig. 2.19, Chemistry in Context

Montreal Protocol Had Banned Most Industrial Production of CFCs & Other ODS

The New York Times

In a High-Stakes Environmental Whodunit, Many Clues Point to China

Interviews, documents and advertisements collected by The New York Times and independent investigators indicate that a major source — possibly the overwhelming one — is factories in China that have ignored a global ban and kept making or using the chemical, CFC-11, mostly to produce foam insulation for refrigerators and buildings.

"You had a choice: Choose the cheaper foam agent that's not so good for the environment, or the expensive one that's better for the environment," said Zhang Wenbo, owner of a refrigerator factory here in Xingfu, in Shandong Province, where he and many other small-scale manufacturers said that until recently, they had used CFC-11 widely to make foam insulation.

Billboards in Xingfu, China, promoting locally made refrigerators. The city has around 1,700 businesses involved in the production of cooking and refrigeration equipment. Gilles Sabrié for The New York Times

https://www.nytimes.com/2018/06/24/world/asia/china-ozone-cfc.html

CFC-11 Emissions Update

Figure ES-3. CFC-11 global emissions and reported production. Shown are emissions of CFC-11 derived from AGAGE (Advanced Global Atmospheric Gases Experiment; red) and NOAA (National Oceanic and Atmospheric Administration; blue) global network measurements of CFC-11 abundances (see also Figure 1-3 of the Assessment) and a model using a CFC-11 lifetime of 52 years. Also shown is the production history reported to the UN Environment Programme for all uses (green), the average of annual emissions over the 2002–2012 period (horizontal purple line) extended to 2020 (dashed purple line), and scenario projections based on observations through 2006 or through 2012 (grey dotted and dashed lines). These emission projections are calculated using standard methodologies based on reported production, inventory estimates of the bank, and an empirically determined release fraction from the bank over the seven years before 2006 or 2012, which is then applied to subsequent years (see Chapters 1 and 7). Uncertainties in emissions, shown as vertical lines on the data points, include the influence of measurement and model representation uncertainties, and do not include the influence of dynamical variability. The uncertainties are smaller than those presented in Figure 1-3, because uncertainties related to factors constant across the whole time period, such as lifetimes and calibration scale, have been omitted.

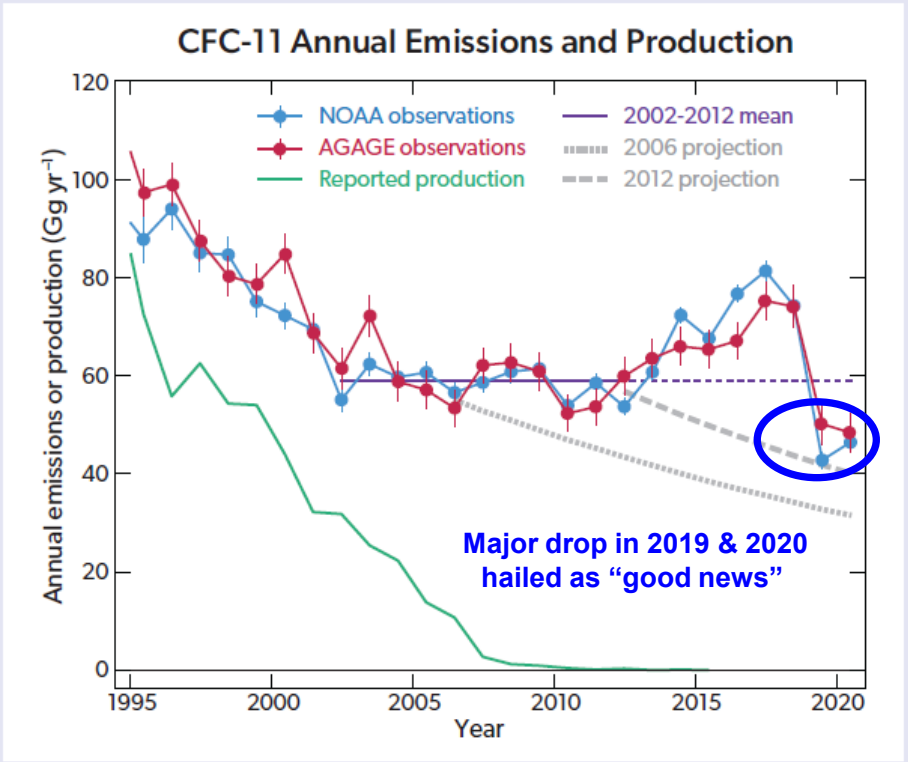
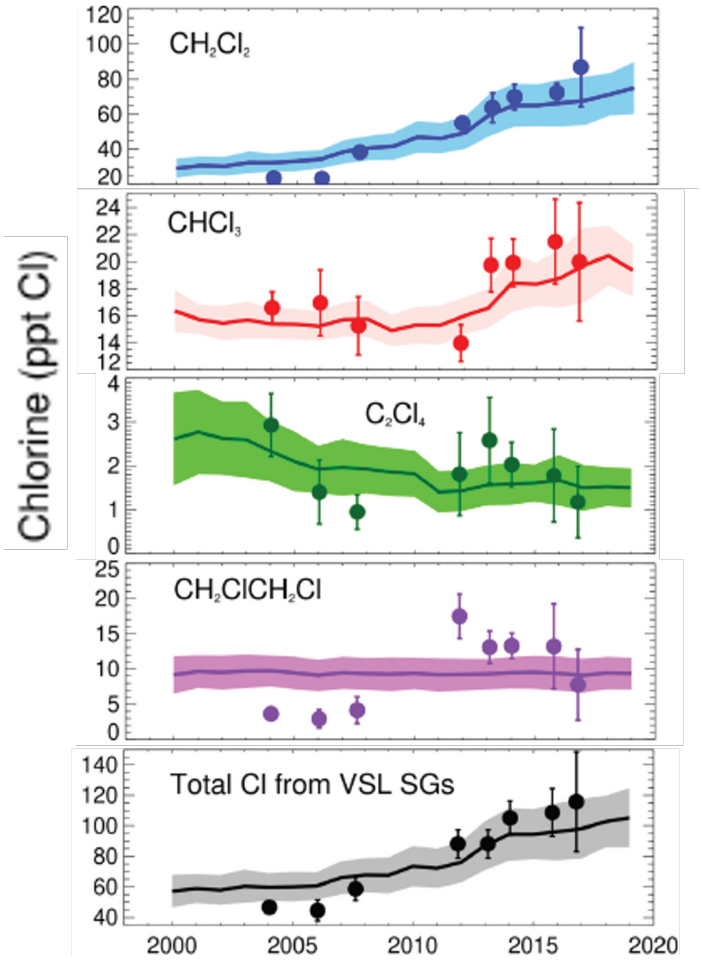


Fig ES-3, 2022 *WMO/UNEP Scientific Assessment of Ozone Depletion Executive Summary*
https://csl.noaa.gov/assessments/ozone/2022/downloads/executivesummary.pdf

Very-Short Lived (VSL) Chlorine

These gases, all mainly anthropogenic, have lifetimes for atmospheric removal of less than 6 months and are not controlled by the Montreal Protocol.



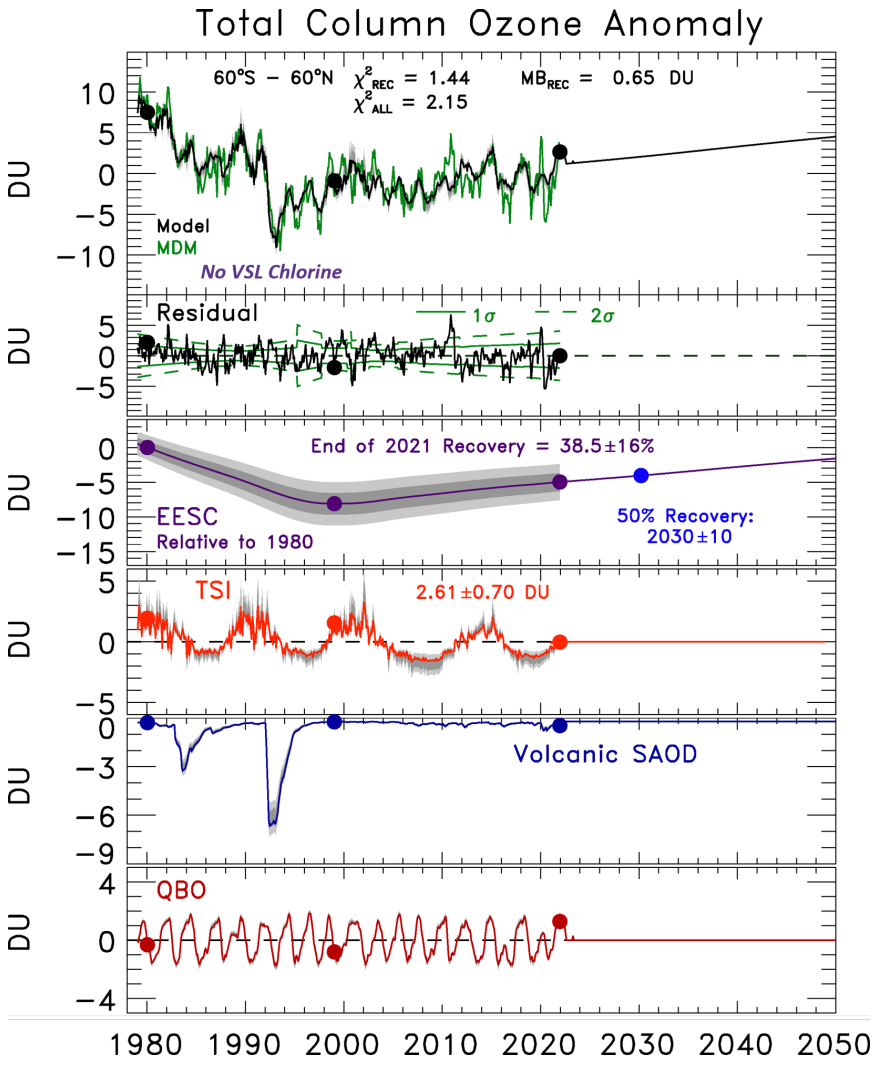
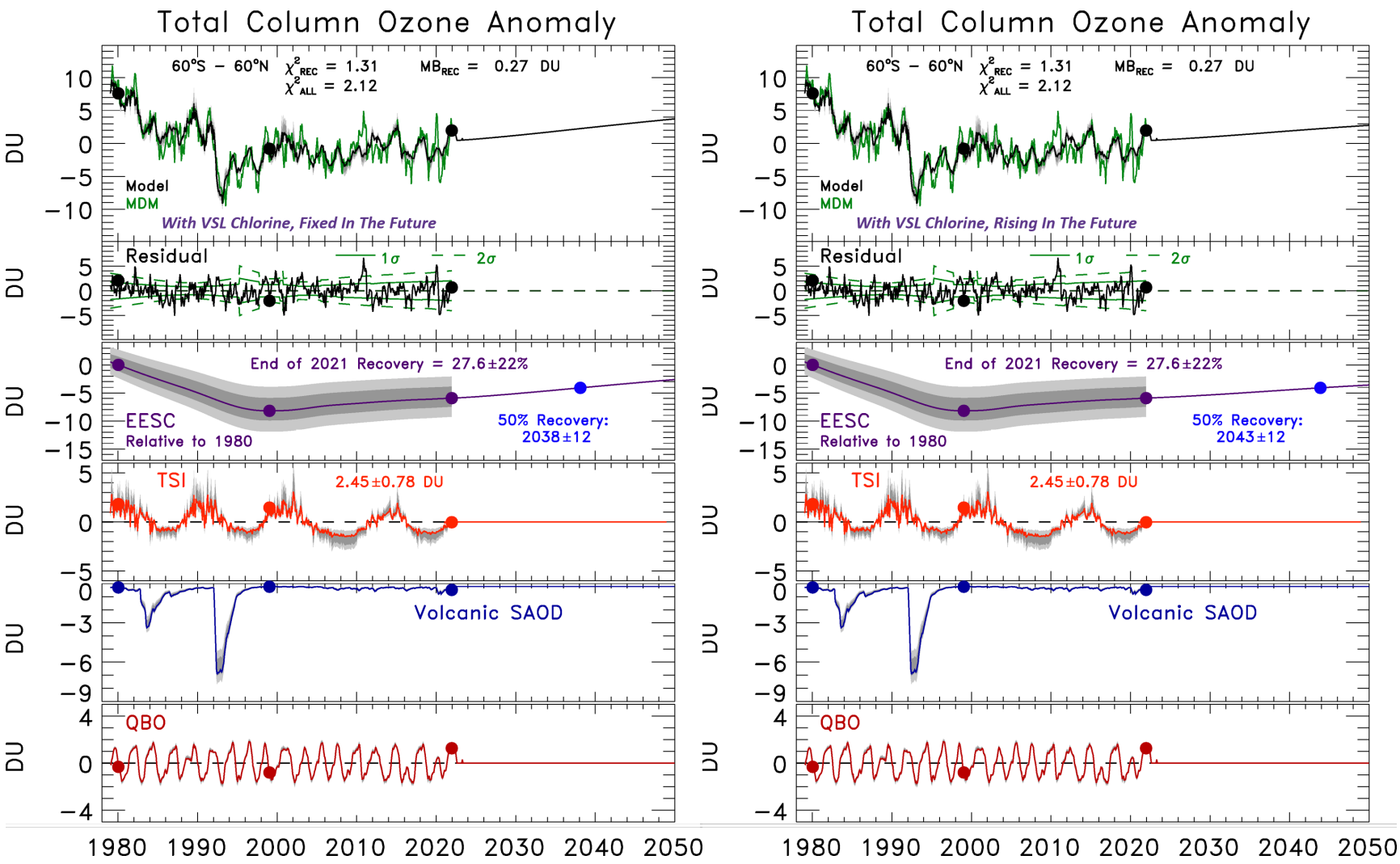
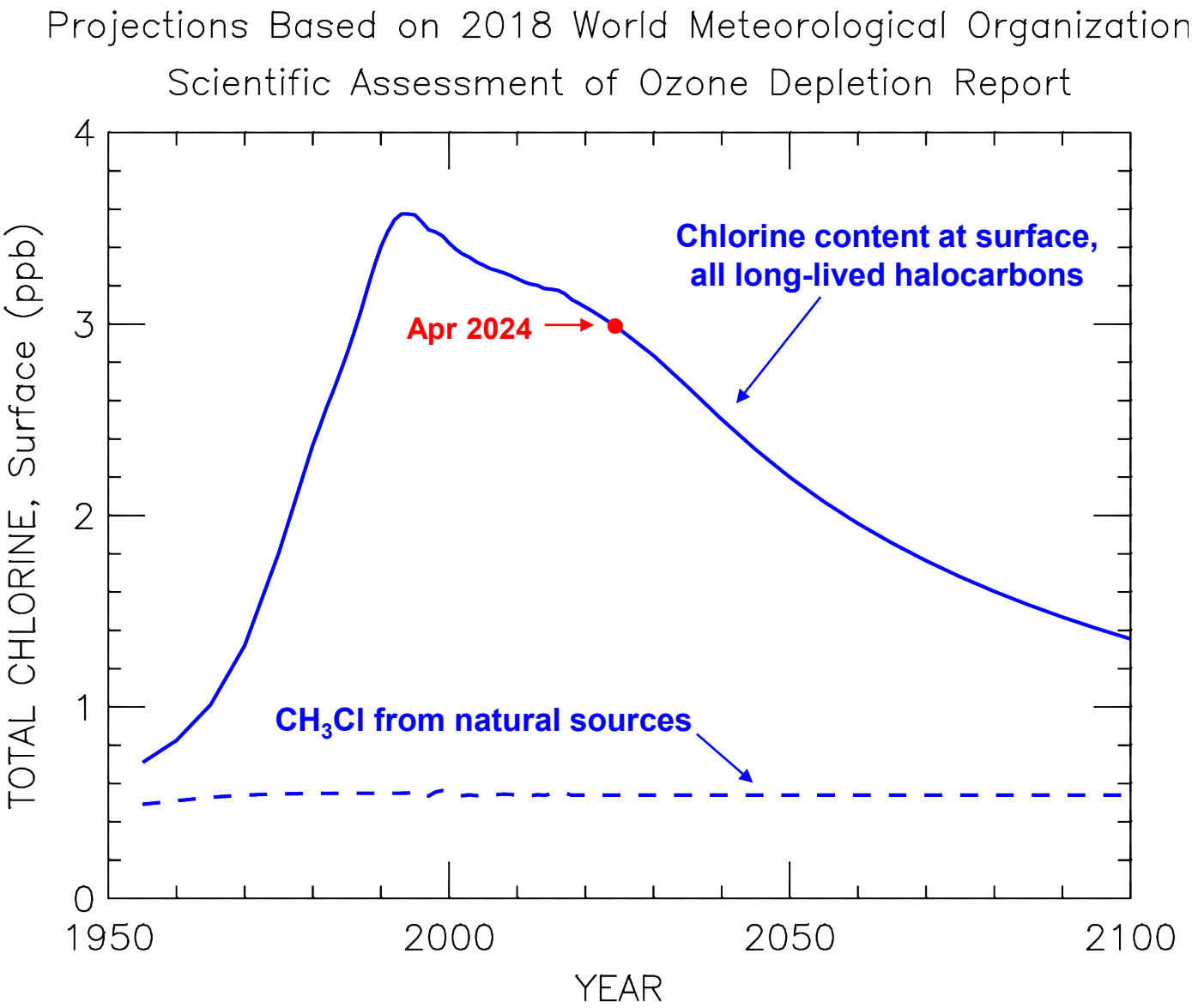


Fig 1-8, 2022 *WMO/UNEP Scientific Assessment of Ozone Depletion Report*
https://csl.noaa.gov/assessments/ozone/2022/downloads/Chapter1_2022OzoneAssessment.pdf

Very-Short Lived Chlorine

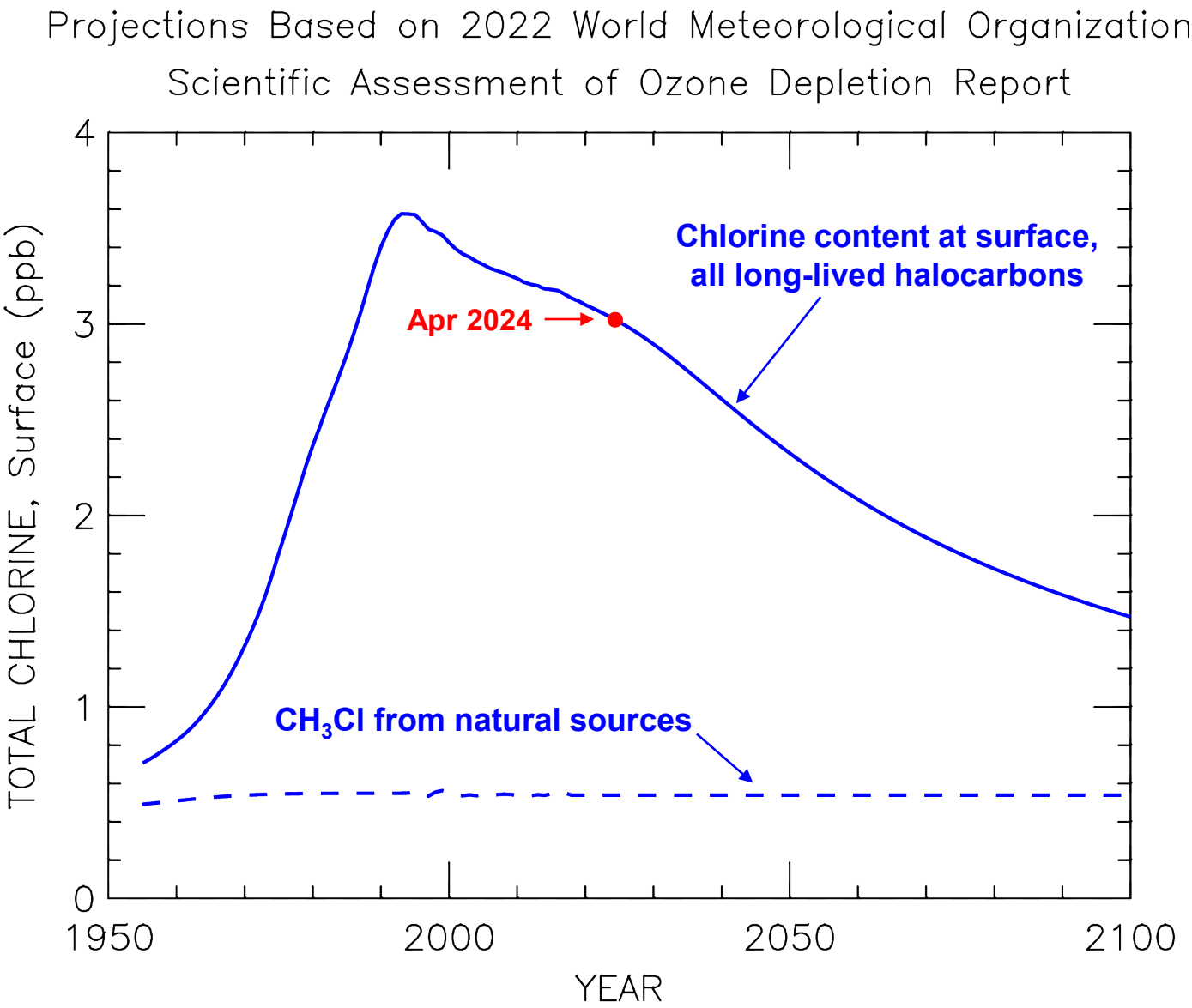


Argh, "Long-Lived" Chlorine Not Declining As Fast As We Had Projected



2018 WMO Scientific Assessment of Ozone Depletion Report:
https://www.esrl.noaa.gov/csd/assessments/ozone/2018

Argh, “Long-Lived” Chlorine Not Declining As Fast As We Had Projected



2022 WMO Scientific Assessment of Ozone Depletion Report:
https://www.esrl.noaa.gov/csd/assessments/ozone/2022

Argh, “Long-Lived” Chlorine Not Declining As Fast As We Had Projected

AMERICAN METEOROLOGICAL SOCIETY

104TH ANNUAL MEETING
28 JANUARY-1 FEBRUARY 2024
BALTIMORE, MD & ONLINE

12A.1 - Kicking the Can Down the Road in Ozone Recovery

Wednesday, January 31, 2024
4:30 PM - 4:45 PM
310 (The Baltimore Convention Center)

***Megan Lickley**, Georgetown University, Washington, DC; Georgetown University, Cambridge, MA; and R. J. Salawitch, J. S. Daniel, L. McBride, and G. Velders*

310 (The Baltimore Convention Center)

Abstract

Every four years the international scientific assessment of ozone depletion is prepared to support decisions made by the Parties to the Montreal Protocol. A key component of each assessment is an outlook of the ozone recovery timeline, which has been quantified using equivalent effective stratospheric chlorine (EESC), a metric that has been developed to relate surface level atmospheric abundance of ozone depleting substances (ODSs) to stratospheric ozone destruction. In each assessment, the year in which EESC values will return to below 1980 levels is estimated, given the best scientific understanding of atmospheric processes and assuming global compliance with the Protocol. However, since 2006, the expected EESC return date to below 1980 levels has been consistently delayed between assessments from an expected EESC return date of 2049 in the 2006 assessment to 2066 in the 2022 assessment, an ozone recovery delay of 17 years over a 16-year assessment period. Has this delay in expected ozone recovery been a result of consistently underestimating global production and emissions of ODSs or due to changes in the scientific understanding and representation of atmospheric processes? Here, we investigate this question by identifying the primary drivers that have delayed the expected ozone recovery date between each consecutive international ozone assessment from 2006 to 2022. We find that changes in the formulation of fractional release factors that underlie the calculation of EESC can only partially explain this delay, and that changes in 1) atmospheric lifetime assumptions 2) bank calculation methods 3) updated historical mole fraction estimates and 4) an under-estimate of the atmospheric release of CCl4 account for much of the remaining delay. Since some of these factors are amenable to future controls (i.e., capture of ODSs from banks and limitations on future atmospheric release of CCl4), it is important to understand the reasons for the delays in the expected ozone recovery date.

https://ams.confex.com/ams/104ANNUAL/meetingapp.cgi/Session/67035