Stratospheric Chemistry: Polar Ozone Depletion

AOSC/CHEM 433 & AOSC/CHEM 633

Ross Salawitch

Class Web Sites:
http://www2.atmos.umd.edu/~rjs/class/fall2020
https://myelms.umd.edu/courses/1291919

Today:

- **Processes that govern the formation of the Antarctic ozone hole**
- **Compare & contrast Antarctic and Arctic ozone depletion**
- **Antarctic & Arctic Ozone 2020**

Lecture 16

5 November 2020

Announcements

- Problem Set #3 has been posted:

  https://www2.atmos.umd.edu/~rjs/class/fall2020/problem_sets/ACC_2020_problem_set_03.pdf

  and is due on ~~Tuesday, 10 November~~ Thursday, 12 November. Please get started early.

- Due to "popular demand", this problem set will be completed outside of ELMS: prefer you mail me and Laura McBride (mcbridel@terpmail.umd.edu) either one PDF file (entire P Set) or two PDF files (one per question) when the problem set is complete, with an email subject such as:

  AOSC 433: Problem Set 3 *or* CHEM 633: Problem Set 3 , etc

- Please email me and Laura with questions

- No AT for Lecture 17 on Tuesday, 10 November. However, please try to complete short reading.

Problem Set #3 due date extended to Thursday, 12 Nov due to issues with the class website on 5 Nov 2020.

Announcements: Outside of Class

1) Today, 5 Nov: AOSC Weekly Seminar (3:30 pm)

Dr. Carey King, Energy Institute, University of Texas at Austin

The economic superorganism: Beyond the competing narratives on energy, growth, and policy

Energy drives the economy, economics informs policy, and policy affects social outcomes. Since the oil crises of the 1970s, pundits have debated the validity of this sequence, but most economists and politicians still ignore it. Thus, they delude the public about the underlying influence of energy costs and constraints on economic policies that address such pressing contemporary issues as income inequality, growth, debt, and climate change. To understand why, Carey King will explore the scientific and rhetorical basis of the competing narratives both within and between energy technology and economics.

The competing energy narratives pit "drill, baby, drill!" (fossil fuels) against renewable technologies such as wind and solar. Both claim to provide secure, reliable, clean, and affordable energy to support economic growth with the most benefit to society, but how? To answer this question, we need to understand the competing economic narratives, techno-optimism and techno-realism. Techno-optimism claims that innovation overcomes any physical resource constraints and enables the social outcomes and economic growth we desire. Techno-realism, in contrast, states that no matter what energy technologies we use, feedbacks from physical growth on a finite planet constrain economic growth and create an uneven distribution of social impacts. But we don't need these narratives, and in fact they distract us, to more accurately understand how the physical basis of the economy is affecting social outcomes.

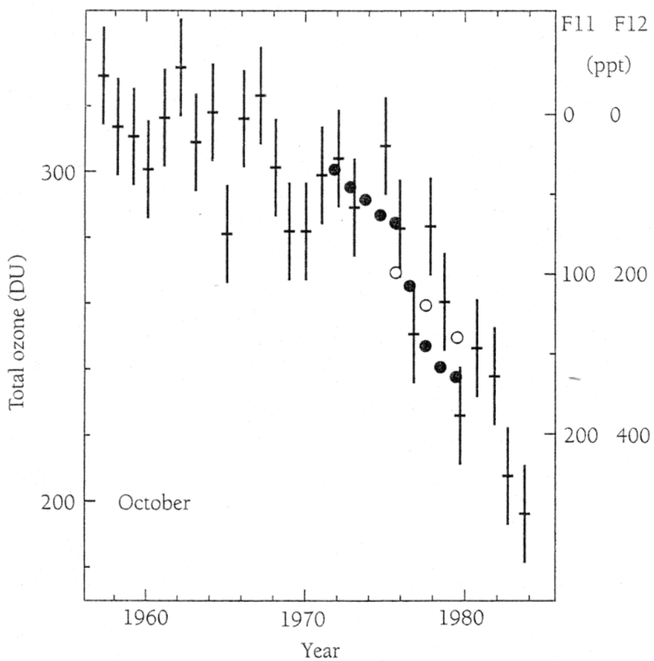
https://aosc.umd.edu/seminars/department-seminar

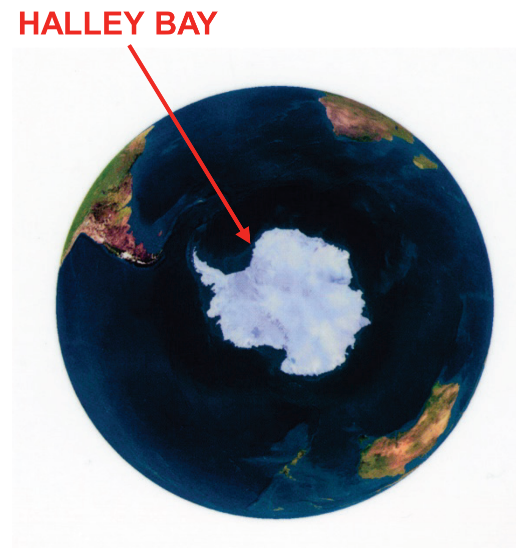
Email Joseph Knisely at jknisely@umd.edu for Zoom connection info

Polar Ozone Depletion

Discovery of the ozone hole:

Halley Bay (76°S)
October Mean



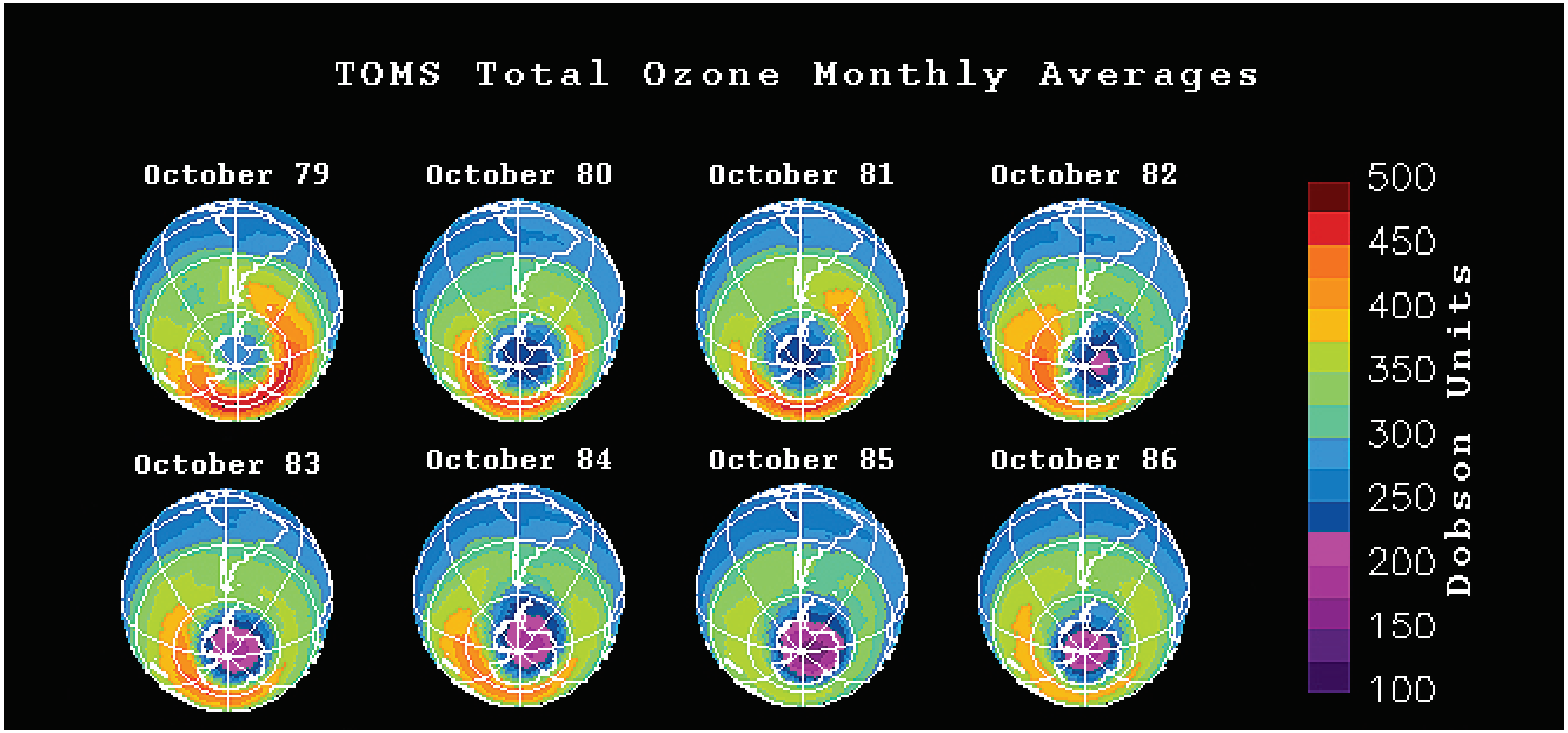


Southern Hemisphere

Farman *et al*., Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction, *Nature, 315*, 207, 1985.

Polar Ozone Depletion

First view from space:



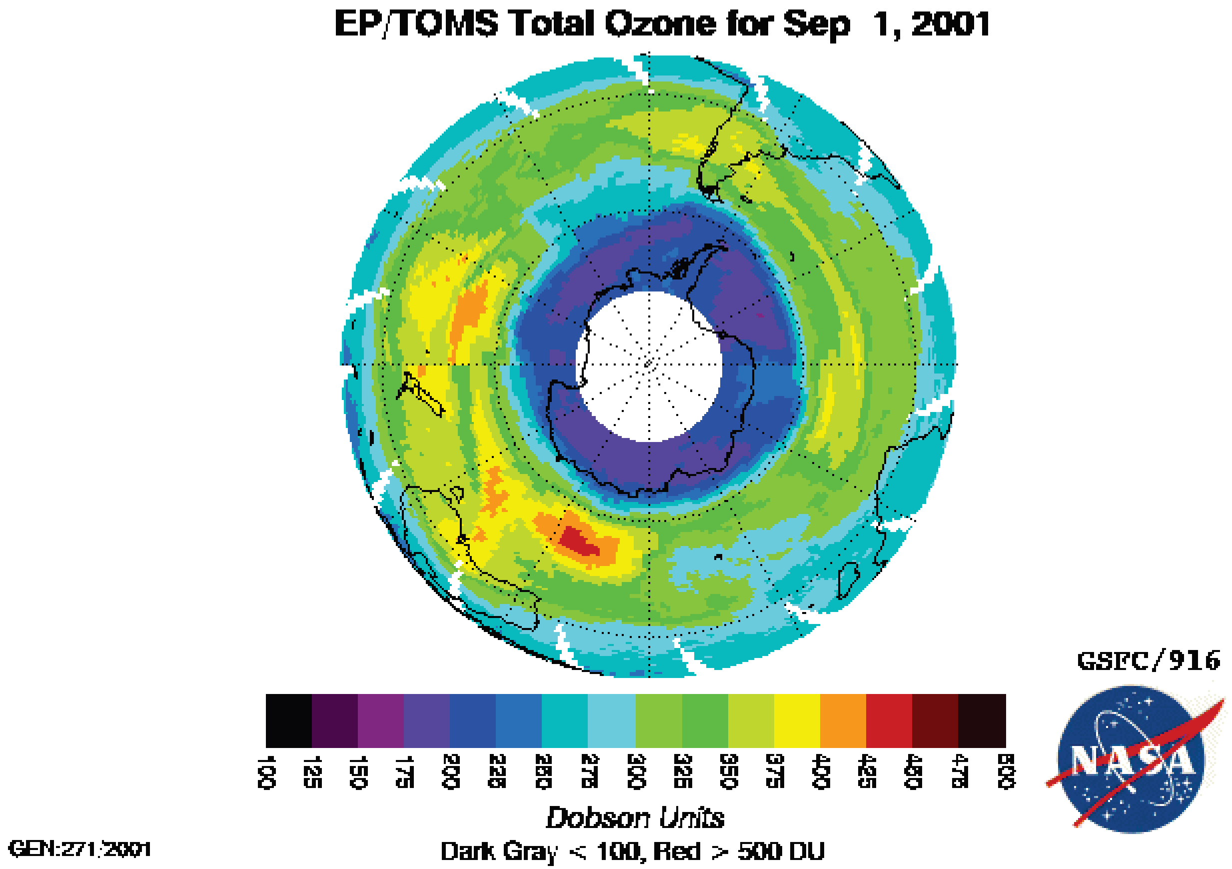
Stolarski *et al*., *Nature*, 322, 808, 1986.

The paper showed data for Octobers of 1979 through 1985 in black & white contour diagrams. This image, produced soon after, showed color plots of total column ozone during Antarctic spring, including measurements for year 1986.

Polar Vortex Circulation

During winter:

- radiative cooling leads to cold air in polar stratosphere
- large scale low pressure region develops over pole
- strong "polar night jet" develops, isolating air at high latitudes from air at low latitudes
- T continues to fall in the "vortex like" circulation near the pole



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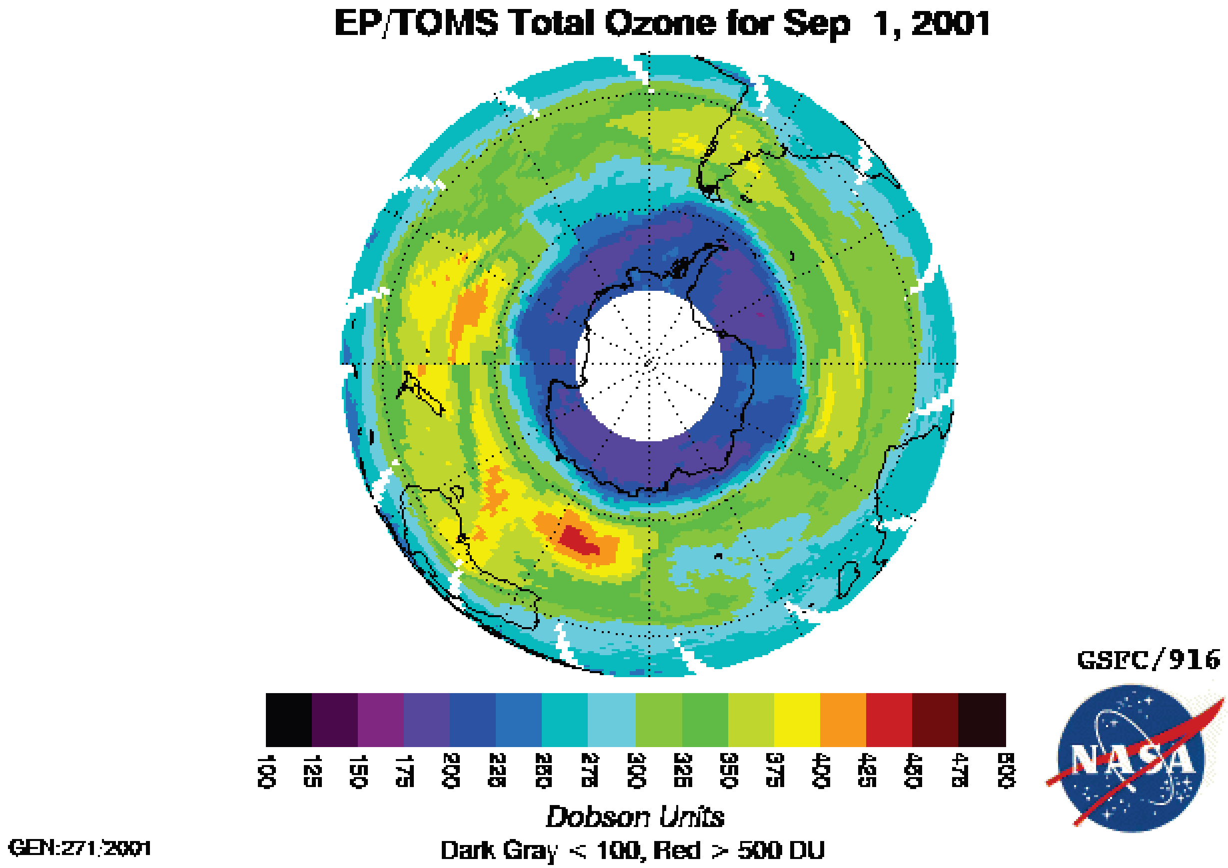


Polar Ozone Depletion Theories

Soon after the discovery of the ozone hole three theories emerged to explain the rapid springtime loss of ozone over Antarctica:

1. Chemistry due to **enhanced levels of ClO**, driven by heterogeneous reactions on the surface of polar stratospheric clouds (PSCs) [McElroy et al., *Nature*, 1986; Solomon et al., *Nature*, 1986]

    a) two new catalytic cycles, both involving halogen radicals and requiring ~1 ppb of ClO to be effective ($ClO + ClO + M \rightarrow ClOOCl + M$; $BrO + ClO \rightarrow Br + Cl + O_2$)
    [Molina and Molina, *JPC*, 1987; McElroy et al., *Nature*, 1986]

    b) suggestion that PSC particles might be composed of HNO_3 and upon sedimentation could appreciably lower NO_x (which would prevent conversion of ClO to $ClNO_3$)
    [Toon et al., *GRL*, 1986]

    c) decreasing ozone column driven by rising ClO, due to buildup of chlorine from CFCs

2. Chemistry due to **enhanced levels of NO_x**, driven by variations in solar UV
   [Callis and Natarajan, *Nature*, 1986]

3. Loss by transport due to **upwelling of ozone poor air** from the troposphere
   [Tung et al., *Nature*, 1986]

Polar Stratospheric Clouds

- Studies prior to the discovery of the ozone hole documented :
  - high altitude (~20 km) "mother of pearl" clouds over Norway
    [e.g., Carl Stormer, Remarkable clouds at high altitudes, Nature, *1929*]
  - greater prevalence of polar stratospheric clouds in SH compared to NH
    [e.g., McCormick et al., Polar Stratospheric Cloud Sightings by SAM II, *JAS*, 1982].

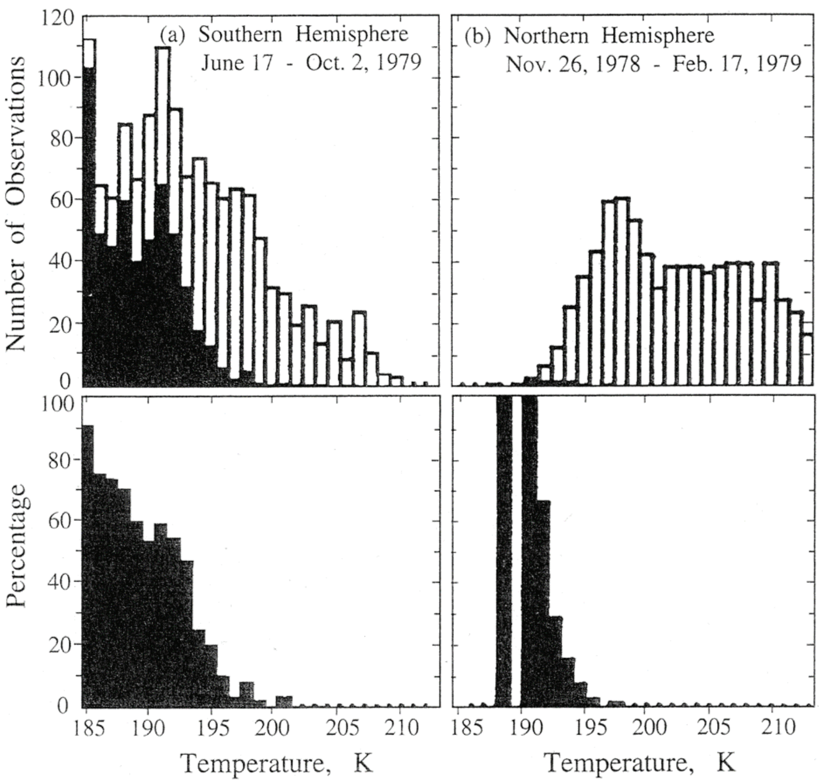
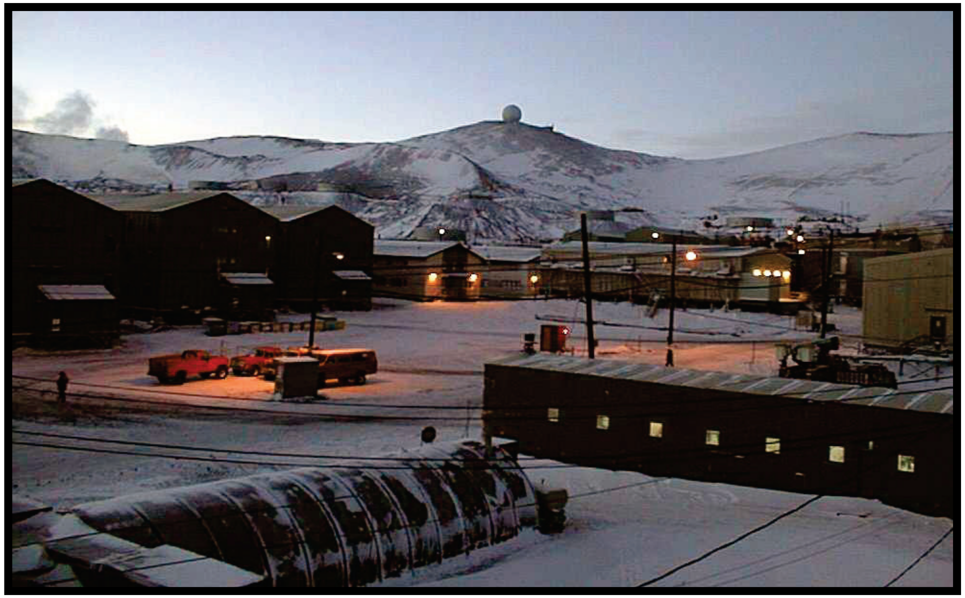


FIGURE 4.20 The top panels show a histogram of the total number of vertical temperature profiles having a given minimum temperature for the Antarctic and Arctic winters. The darkened bars represent observations of PSCs. The lower panel shows the frequency of PSC observations as a percentage of the total events with the same minimum temperature (McCormick et al., 1982). All events for temperatures ≤185 K are included in the 185 K bin.

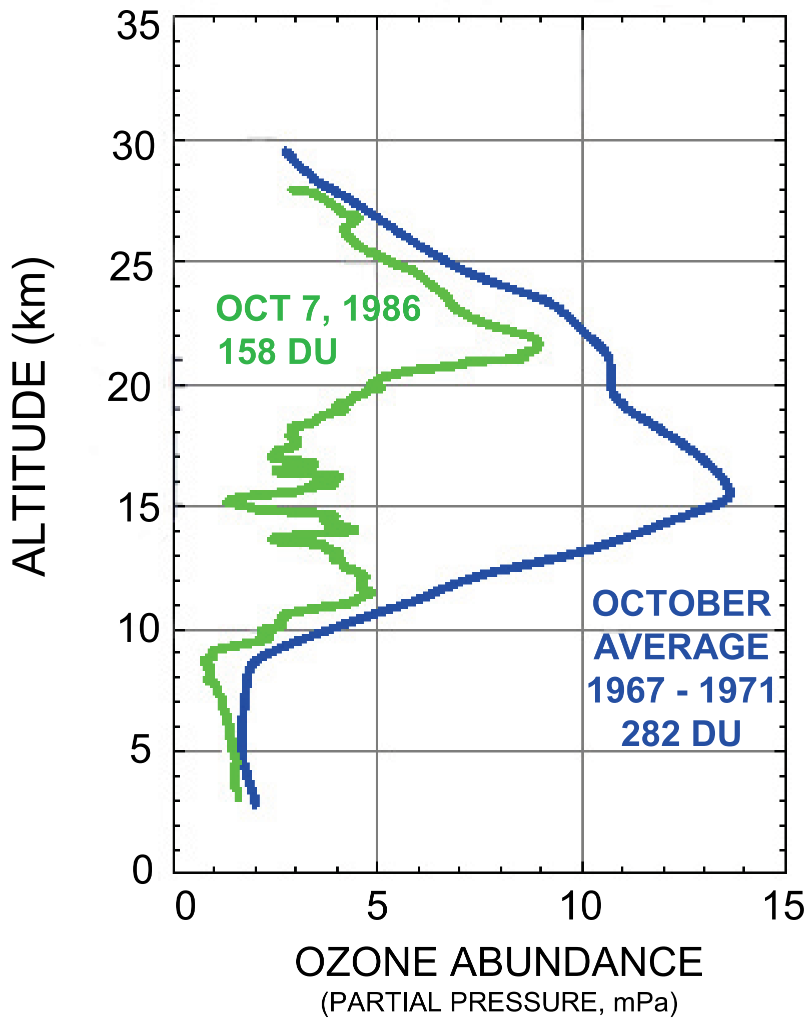
National Ozone Expedition: McMurdo Station, 1986



National Ozone Expedition: McMurdo Station, 1986

Balloon-borne ozonesondes showed:

Region of nearly complete removal of ozone between ~12 and 20 km:

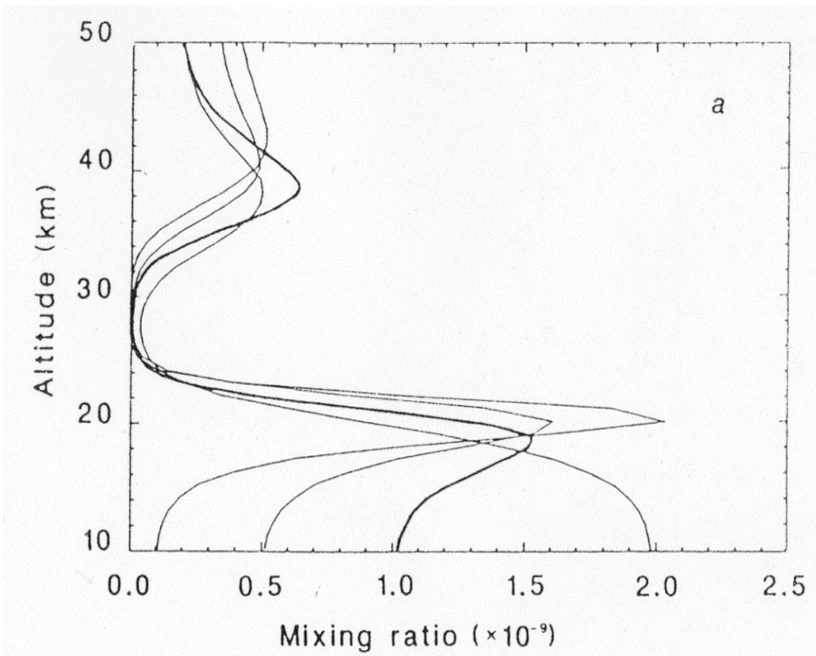


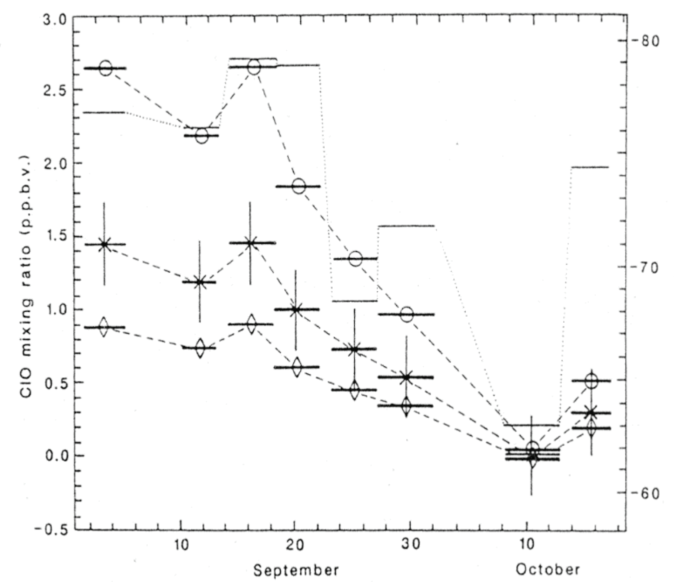
Hofmann et al., *Nature, 326*, 59, 1987.

National Ozone Expedition: McMurdo Station, 1986

Ground based measurements revealed:

- Presence of ~1 ppb of ClO over Antarctica
- Decreasing column HNO_3 throughout springtime and suppressed column HCl and $ClNO_3$, consistent with existence of large amounts of ClO [Farmer et al., *Nature*, 1987]





Left: ClO profiles retrieved over McMurdo Station based on ground based microwave spectra acquired 1-22 Sept 1986, for initial mixing ratio guesses of 0.1, 0.5, 1.0, and 2.0 ppb. Because pressure broadening > spectral bandwidth below ~15 km, the initial guess is unaltered by the retrieval algorithm below ~15 km. ***Right:*** Time series of ClO over McMurdo, assuming constant ClO mixing ratio vs altitude between 15-20 km (circles), 15-22 km (crosses), or 15-24 km (diamonds). Thin lines connected by dots are stratospheric temperature at 18 km. From *DeZafra et al*., *Nature*, 1987 and *P. Solomon et al*., *Nature,* 1987.

Chlorine Abundance, Mid-Latitude Stratosphere

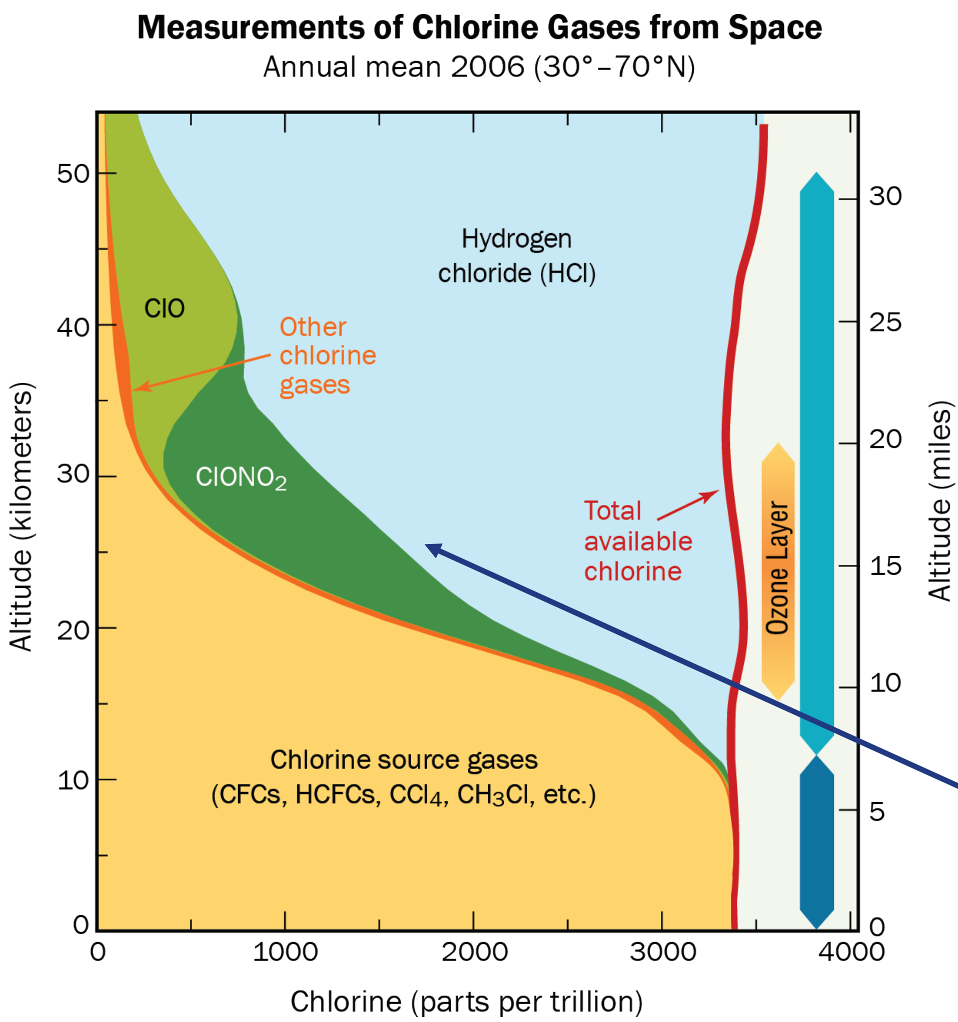
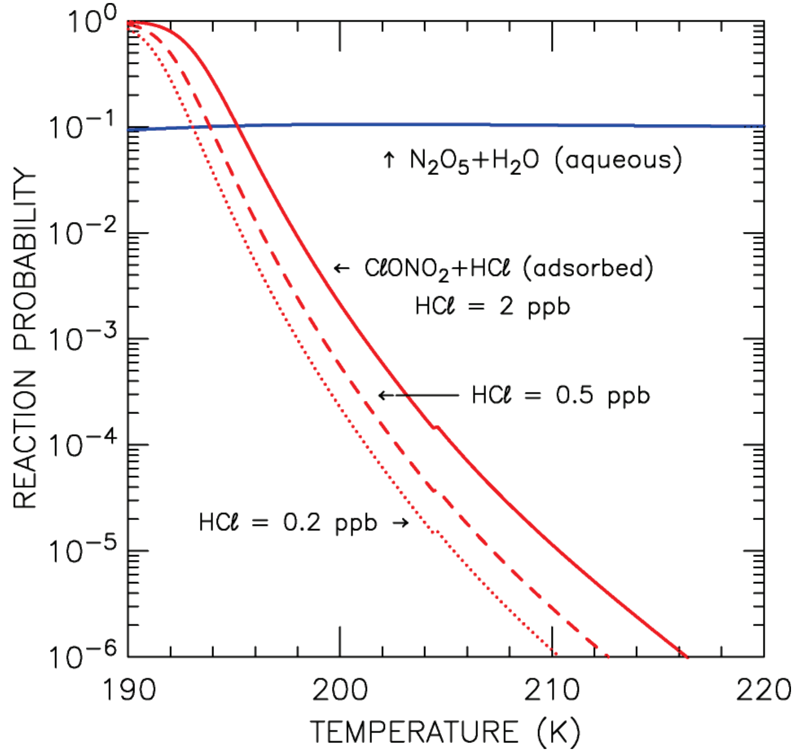


Fig Q8-2, 20 QAs, WMO (2014)

Note: Below ~30 km,
ClO << $ClNO_3$ and HCl

Heterogeneous Chemistry, Mid-Latitude vs Polar Regions

In all cases, γ must be measured in the laboratory



Reaction probabilities given for various surface types, with formulations of various degrees of complexity, in **Section 5** of the JPL Data Evaluation.

Atmospheric Chemistry and Physics by Seinfeld and Pandis provides extensive treatment of aqueous phase chemistry, properties of atmospheric aerosol, organic aerosols, etc.

Heterogeneous Chemistry, Mid-Latitude vs Polar Regions

a) What type of aerosol particles are present in the mid-latitude stratosphere (i.e., what is the chemical composition of these aerosols?)

b) What heterogeneous chemical reaction occurs on the aerosol particles present in the mid-latitude stratosphere and how is the abundance of ClO affected by this reaction?

c) What type of particles are present in the polar stratosphere during winter (i.e., what are these particles called and what is the chemical composition of a "common type" of these particles?)

d) What is the effect of these particles on the chemical composition of the polar stratosphere? Scientists have shown that chemical reactions occurring on the surface of these particles convert species such as and (that do not deplete ozone) and that, while reactive, do not not cause harm to the ozone layer during the darkness of winter.

e) Following the return of sunlight, significant levels of what radical compound builds up inside the Antarctic stratosphere, leading to rapid loss of ozone?

f) Why does the ozone hole occur only over Antarctica?

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Antarctic winter polar vortex is only place in Earth's stratosphere that consistently cools below formation threshold of polar stratospheric

Polar Ozone Loss

- Cold Temperatures → Polar Stratospheric Clouds (**PSCs**)
- Reactions on PSC surfaces lead to elevated **ClO**

  $HCl + ClNO_3 \rightarrow Cl_2$ (gas) + HNO_3 (solid)

  $ClNO_3 + H_2O \rightarrow HOCl + HNO_3$

  Cl_2 + SUNLIGHT + $O_3 \rightarrow ClO$

  HOCl + SUNLIGHT + $O_3 \rightarrow ClO$

  HNO_3 SEDIMENTS (PSCs fall due to gravity)
- Elevated **ClO** + Sunlight destroys O_3
- BrO : Reaction Partner For ClO ⇒ Additional O_3 Loss


N817NA


809
NASA

Roll the Tape

35:15 to 36:40

https://www.facebook.com/nasaearth/videos/live-now-watch-nasas-high-flying-er-2-aircraft-take-off-from-marine-corps-base-h/10155077063157139/

https://www.youtube.com/watch?v=inEQBAzWX_M

0:35 to 2:56

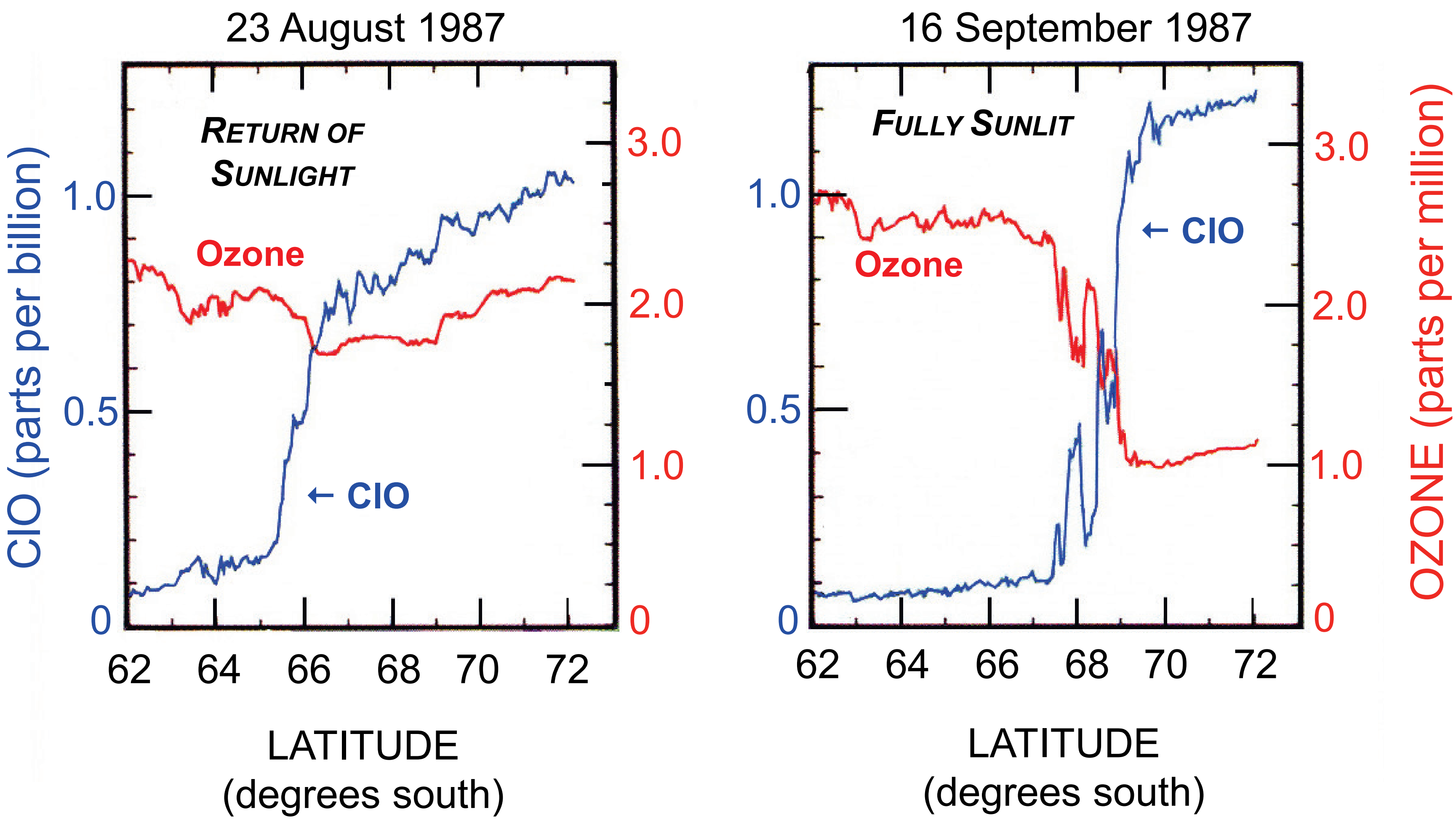
Bonus

NASA ER-2 Takeoff
Created by **MAXAerospace** · Jan 29, 2017

https://coub.com/view/ri2fx

To learn more about the NASA ER-2, which flies at the edge of outer space check out this web-page:
https://video.wired.com/watch/flight-lab-suit-up-and-fly-high-in-nasa-s-science-spy-plane

Airborne Antarctic Ozone Expedition: Punta Arenas, Chile,1987



Anderson et al., *Science*, 1991

Airborne Antarctic Ozone Expedition:

Anderson et al., *Science*, 1991

Polar Ozone Loss Cycles

Cycle (1a):

$ClO + ClO + M \rightarrow ClOOCl + M$

$Cl + O_3 \rightarrow ClO + O_2$

$Cl + O_3 \rightarrow ClO + O_2$

$ClOOCl + h\nu \rightarrow ClOO + Cl$

$ClOO + heat \rightarrow Cl + O_2$

Net: $O_3 + O_3 \rightarrow 3\ O_2$

Cycle (1b):

$ClO + ClO + M \rightarrow ClOOCl + M$

$ClOOCl + heat \rightarrow ClO + ClO$

Net: $M + heat \rightarrow M$

Cycle (2a):

$BrO + ClO \rightarrow BrCl + O_2$

$Br + O_3 \rightarrow BrO + O_2$

$Cl + O_3 \rightarrow ClO + O_2$

$BrCl + h\nu \rightarrow Br + Cl$

Net: $O_3 + O_3 \rightarrow 3\ O_2$

Cycle (2b):

$BrO + ClO \rightarrow ClOO + Br$

$Br + O_3 \rightarrow BrO + O_2$

$Cl + O_3 \rightarrow ClO + O_2$

$ClOO + heat \rightarrow Cl + O_2$

Net: $O_3 + O_3 \rightarrow 3\ O_2$

Cycle (2c):

$BrO + ClO \rightarrow OClO + Br$

$OClO + h\nu \rightarrow O + ClO$

$Br + O_3 \rightarrow BrO + O_2$

Net: $O_3 \rightarrow O + O_2$

Cycle (1) accounts for ~60% of polar ozone loss; Cycle (2) accounts for nearly all of the rest

Rate constants and products for these reactions worked out by many scientists:

Molina and Molina, JPC, 1987

Sander, Friedl, and Yung, *Science,* 1989

Moore, Okumura *et al., Phys. Chem. A,*1999

Bloss, Nickolaisen, Sander *et al*., JPC, 2001

Thermal Decomposition

30.5 kcal/mole 2×24.3 kcal/mole

ClOOCl + M → ClO + ClO + M ΔH = 18.1 kcal/mole

$$\frac{k_{\text{THERMAL}}}{k_{\text{FORMATION}}} = e^{(G_{\text{REACTANTS}} - G_{\text{PRODUCTS}})/RT} = K^{\text{EQUILIBRIUM}}$$

JPL Data Evaluation gives values of $K^{\text{EQUILBRIUM}}$ and $k_{\text{FORMATION}}$

$$K^{\text{EQ}} = 1.27 \times 10^{-27}\, e^{(8744/T)}\ \text{cm}^{-3}$$

In equilibrium:

$$k^{\text{THERMAL}}\,[\text{ClOOCl}] = k^{\text{FORMATION}}\,[\text{ClO}]\,[\text{ClO}]$$

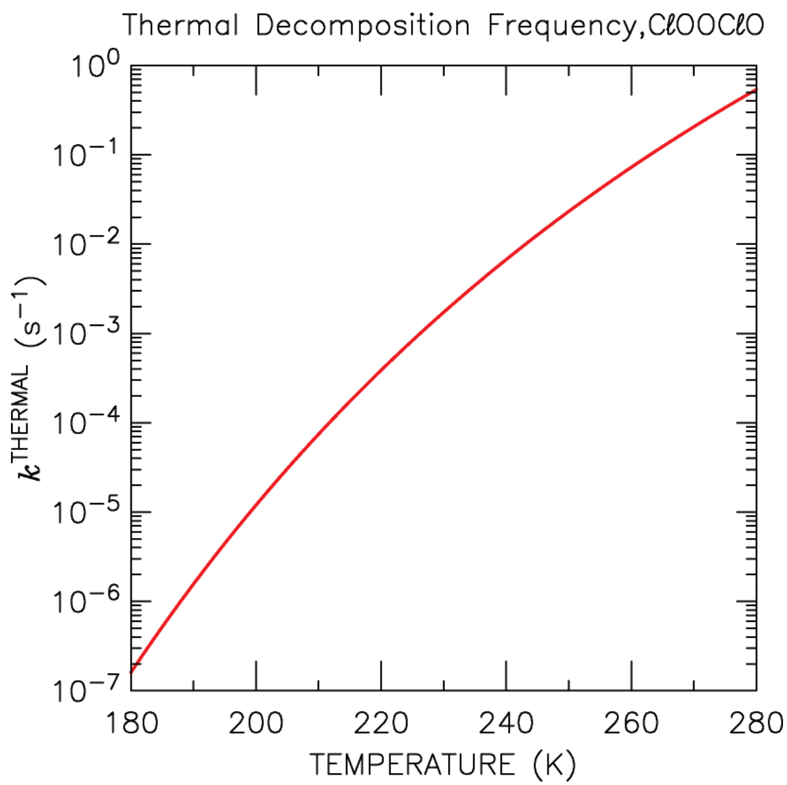
where $k^{\text{THERMAL}} = k^{\text{FORMATION}} \times K^{\text{EQ}}$

- **Energetically, system favors ClOOCl**
- **Entropically, system favors ClO & ClO**

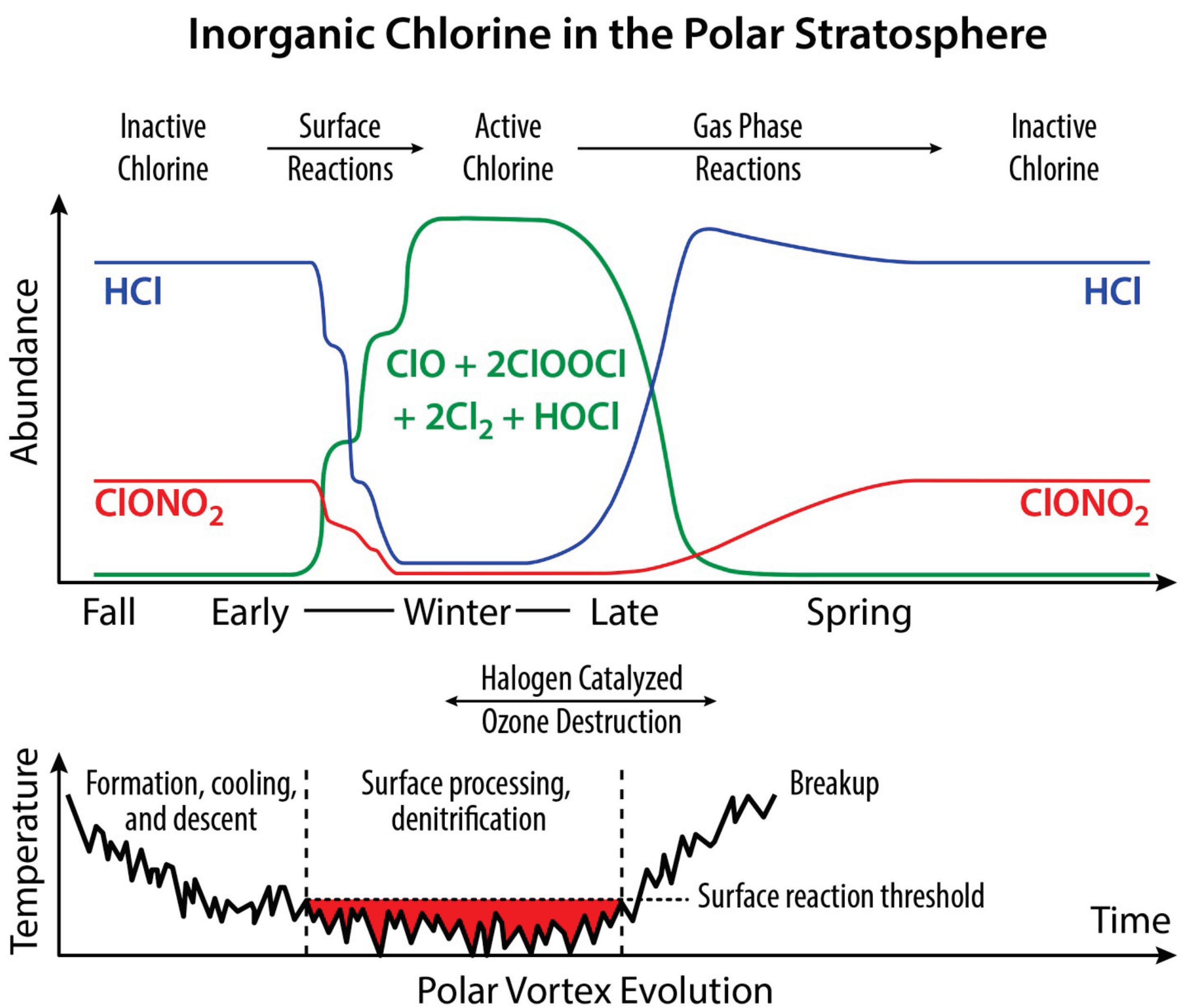
at low T, ClOOCl stable: energy wins !

at high T, ClOOCl unstable: entropy rules !

Equilibrium constants given in **Section 3** of the JPL Data Evaluation.

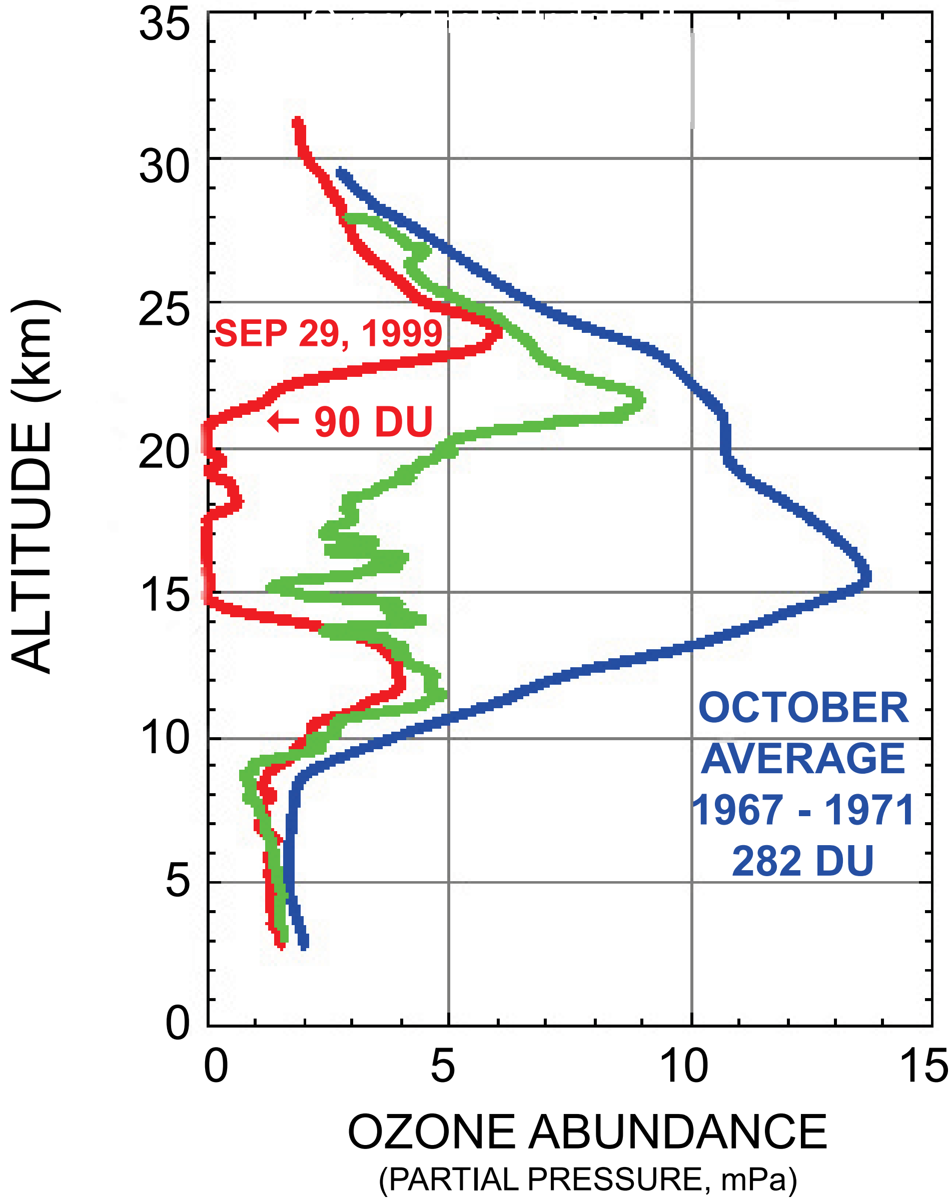


Polar Halogens, Seasonal Evolution



From Wilmouth, Salawitch & Canty, Stratospheric Ozone Depletion and Recovery, 2017

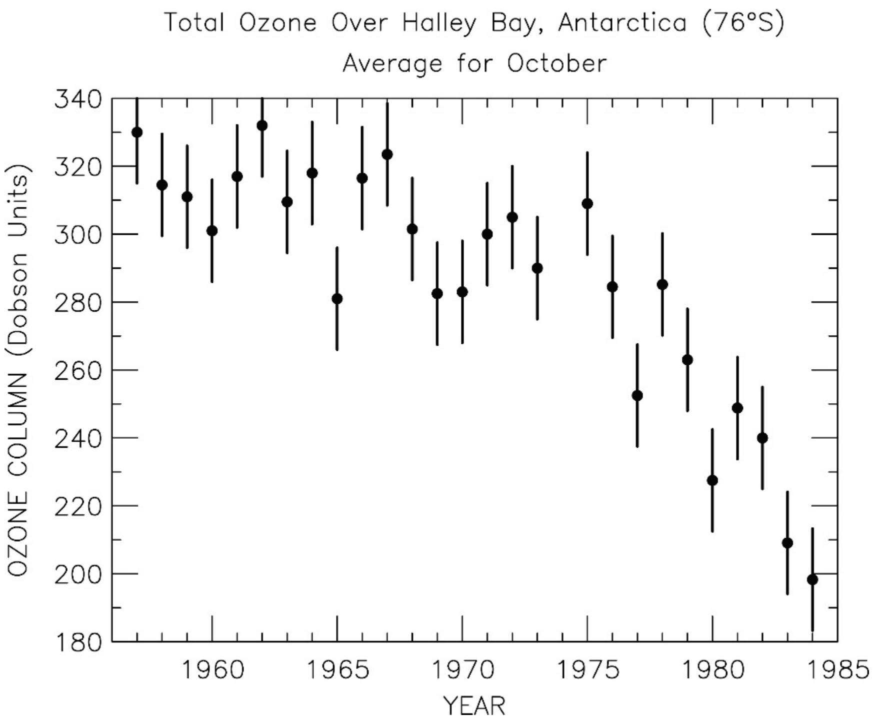
Ozone Profiles, South Pole: Update



D. Hofmann, NOAA CMDL

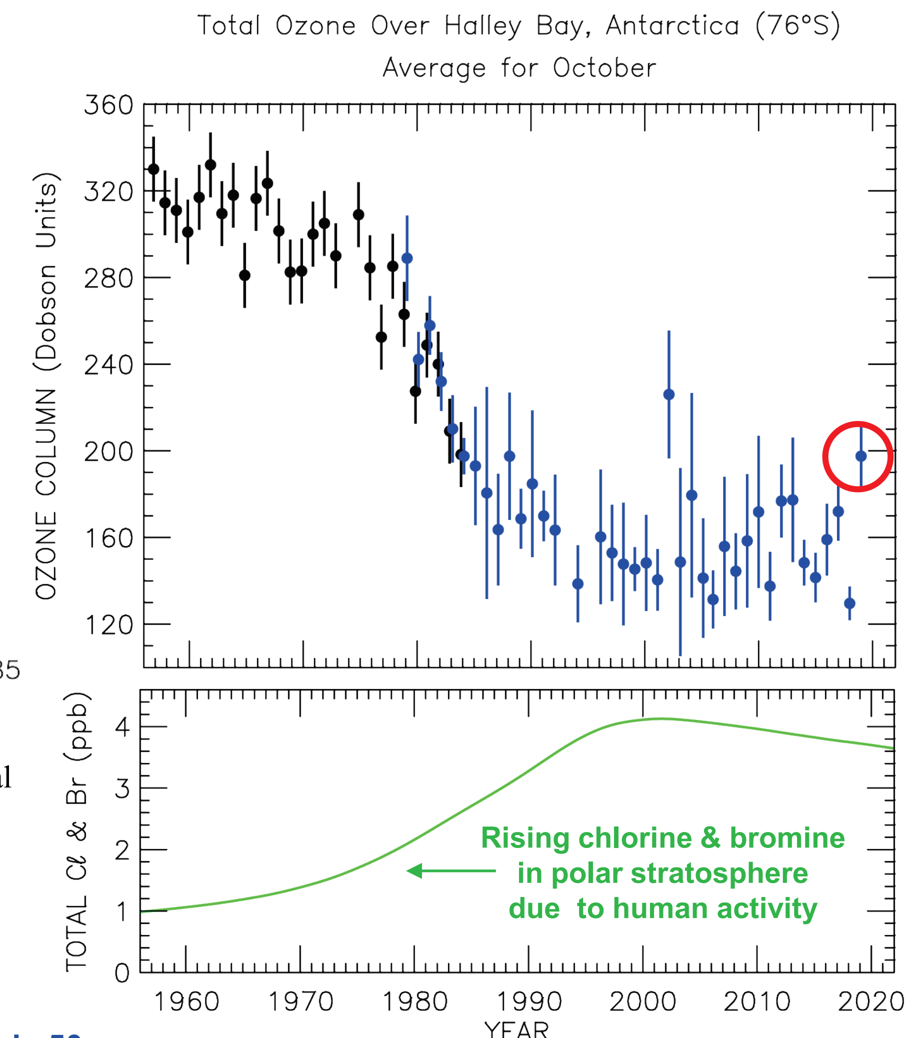
Earth's Atmosphere – Effect of Humans

Stratospheric Ozone – shields surface from solar UV radiation



After Farman *et al*., Large losses of total ozone in Antarctica reveal Seasonal ClOx/NOx interaction, Nature, 315, 207, 1985.

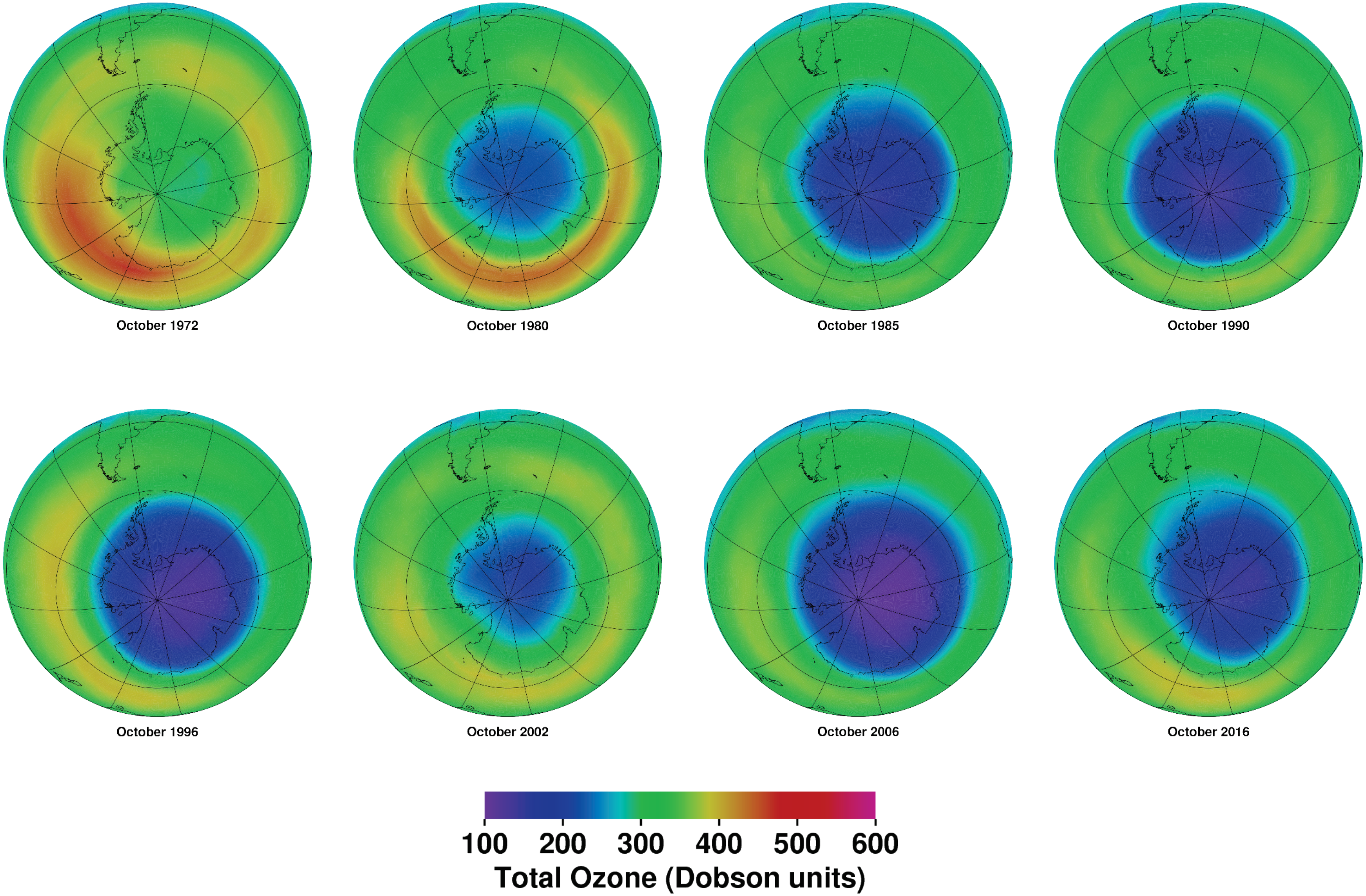
Update



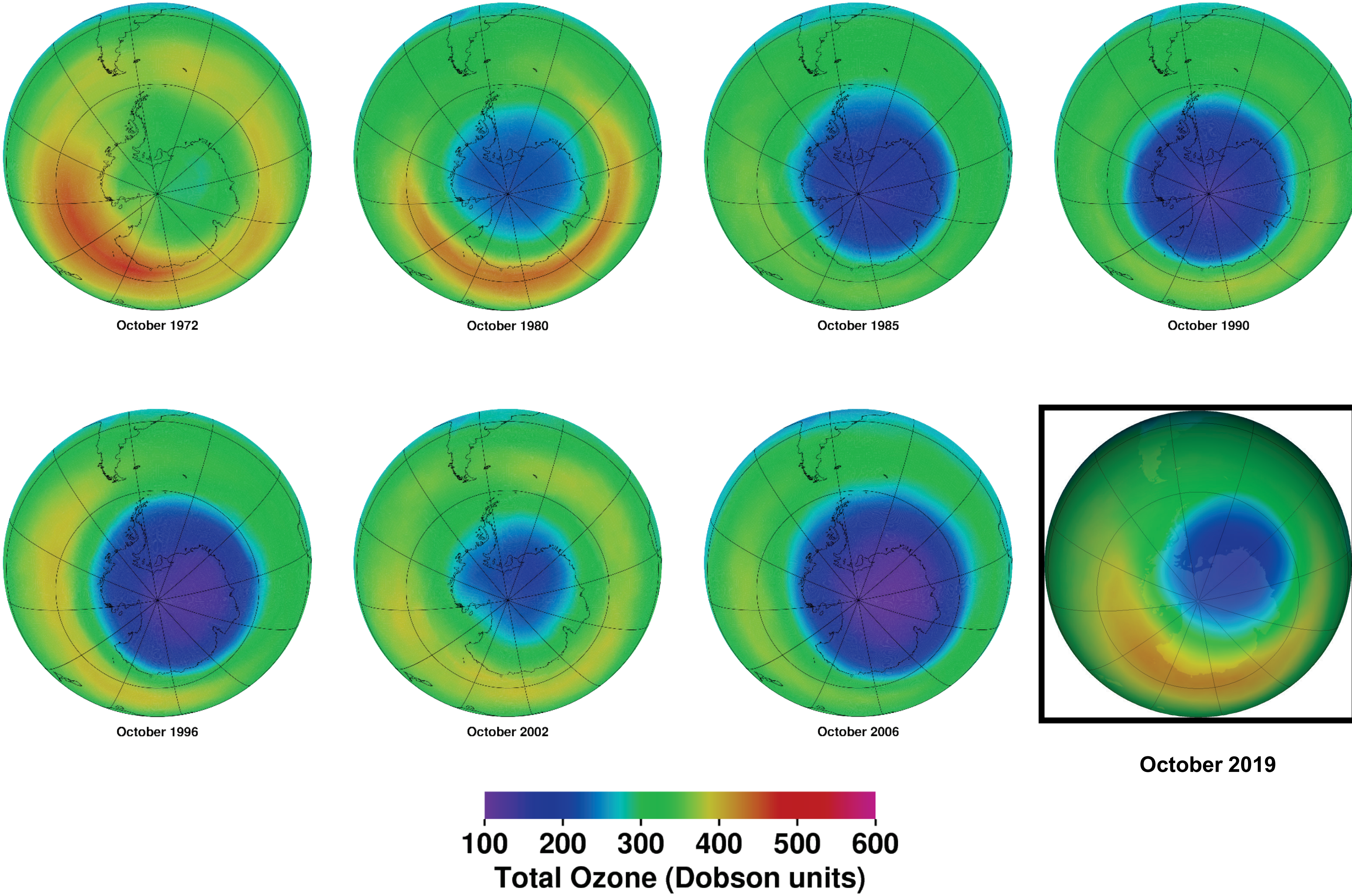
EESC = Total Chlorine + 65 × Total Bromine
(scientists use a multiplier of 65 for polar regions and 60 for extra-polar regions)

Lecture 1, Slide 50

Antarctic Ozone versus Time



Antarctic Ozone versus Time



Antarctic Ozone, 2019

The 2019 Ozone Layer Hole Is Now the Smallest on Record

By Joel Hruska on October 22, 2019 at 3:14 pm | 17 Comments

According to NASA, the ozone layer hole is now the smallest it has been since 1982. The ozone hole is unusually small this year in part because of overall planetary weather patterns that have limited its size. Similar patterns also occurred in 1988 and 2002. NASA is warning people not to expect that recovery has suddenly accelerated as a result of these changes.

"It's great news for ozone in the Southern Hemisphere," said Paul Newman, chief scientist for Earth Sciences at NASA's Goddard Space Flight Center in Greenbelt, Maryland. "But it's important to recognize that what we're seeing this year is due to warmer stratospheric temperatures. It's not a sign that atmospheric ozone is suddenly on a fast track to recovery."

https://www.extremetech.com/extreme/300644-the-2019-ozone-layer-hole-is-now-the-smallest-on-record

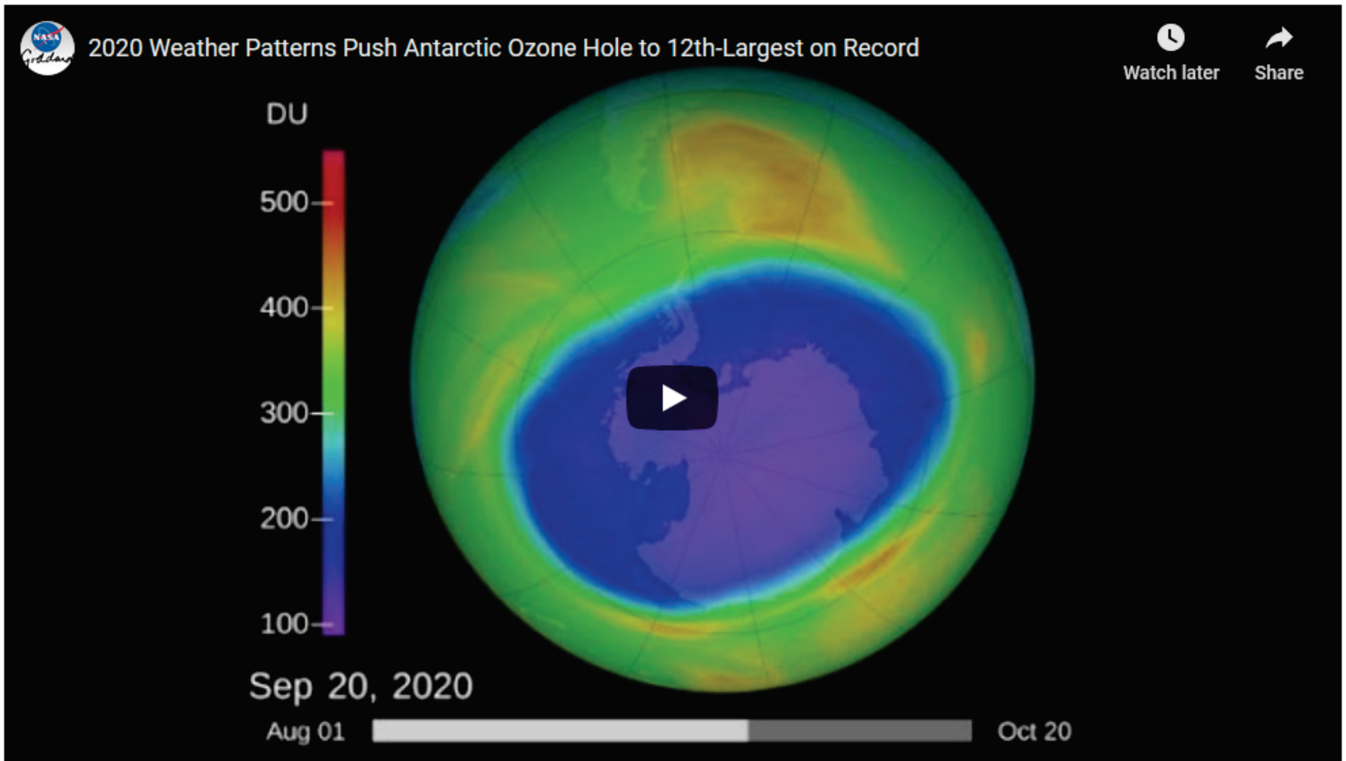
Antarctic Ozone, 2020

Oct. 30, 2020

Large, Deep Antarctic Ozone Hole Persisting into November

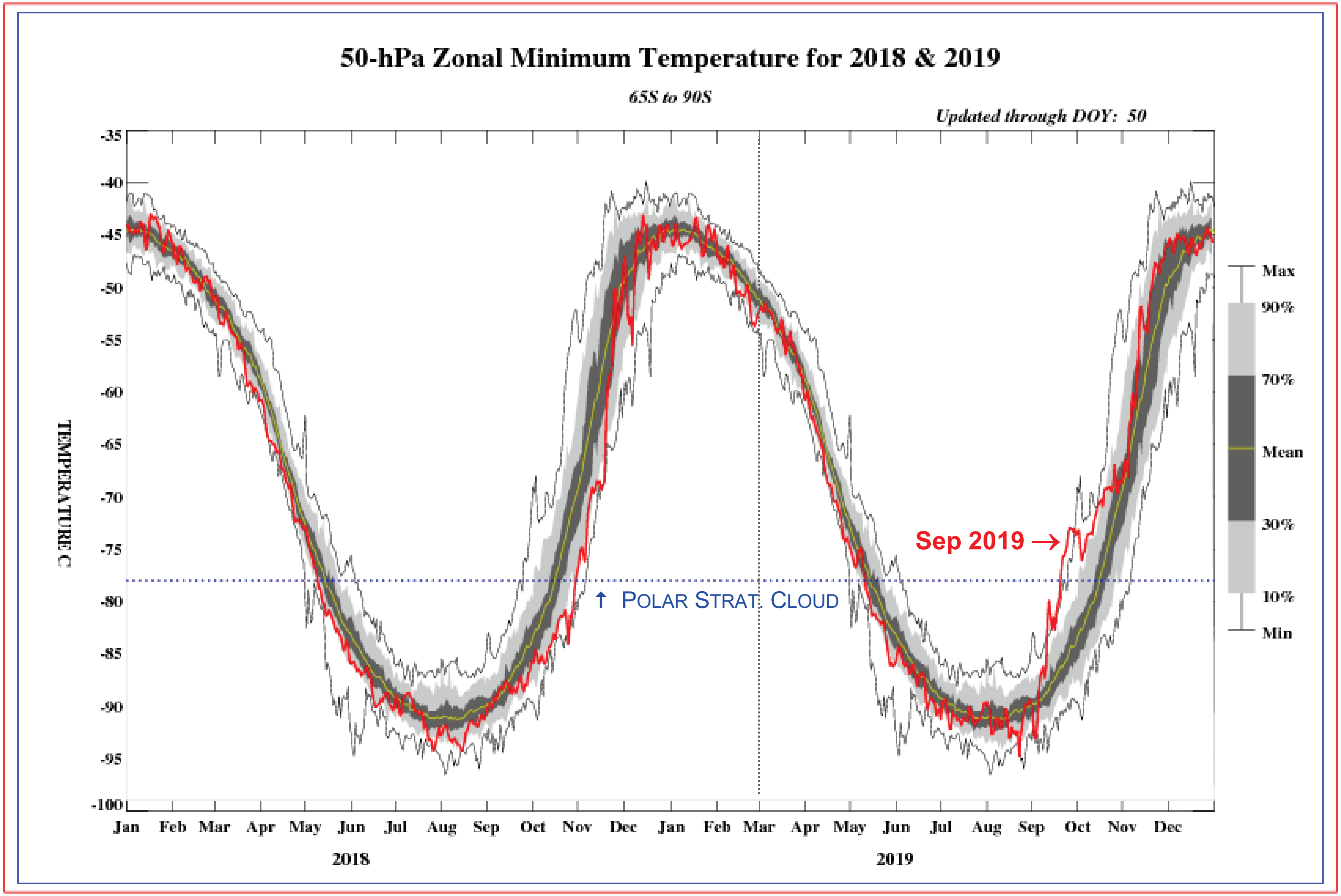
Persistent cold temperatures and strong circumpolar winds, also known as the polar vortex, supported the formation of a large and deep Antarctic ozone hole that should persist into November, NOAA and NASA scientists reported today.

The annual Antarctic ozone hole reached its peak size at about 9.6 million square miles (24.8 million square kilometers), roughly three times the area of the continental United States, on Sept. 20. Observations revealed the nearly complete elimination of ozone in a 4-mile-high column of the stratosphere over the South Pole.



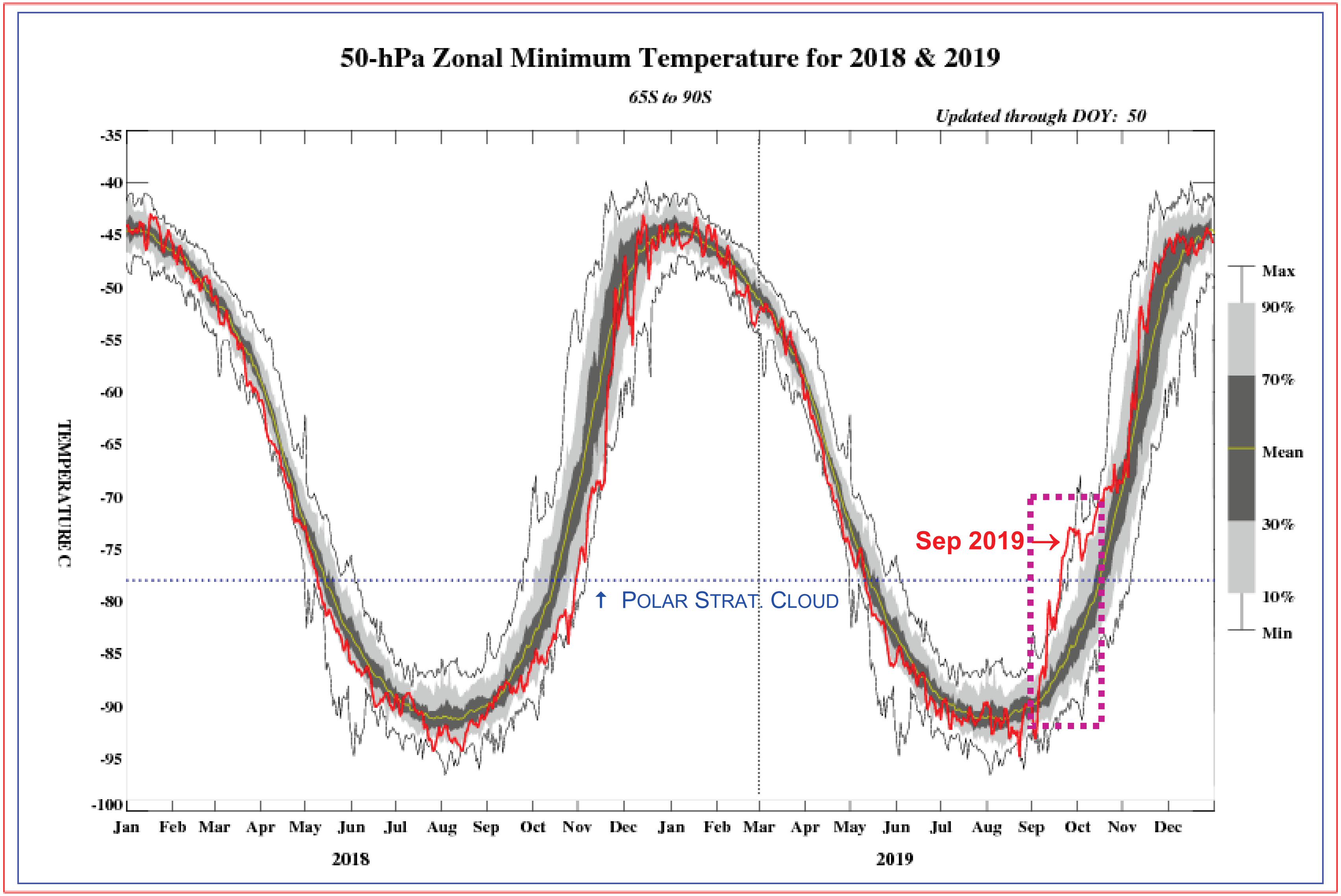
Please watch this 3 minute video: https://www.youtube.com/watch?v=4aq_F9Ma0DQ

Antarctic Vortex Minimum Temperature: 2018-2019



https://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/archive/50mbshlo_2019.png

Antarctic Vortex Minimum Temperature: 2018-2019



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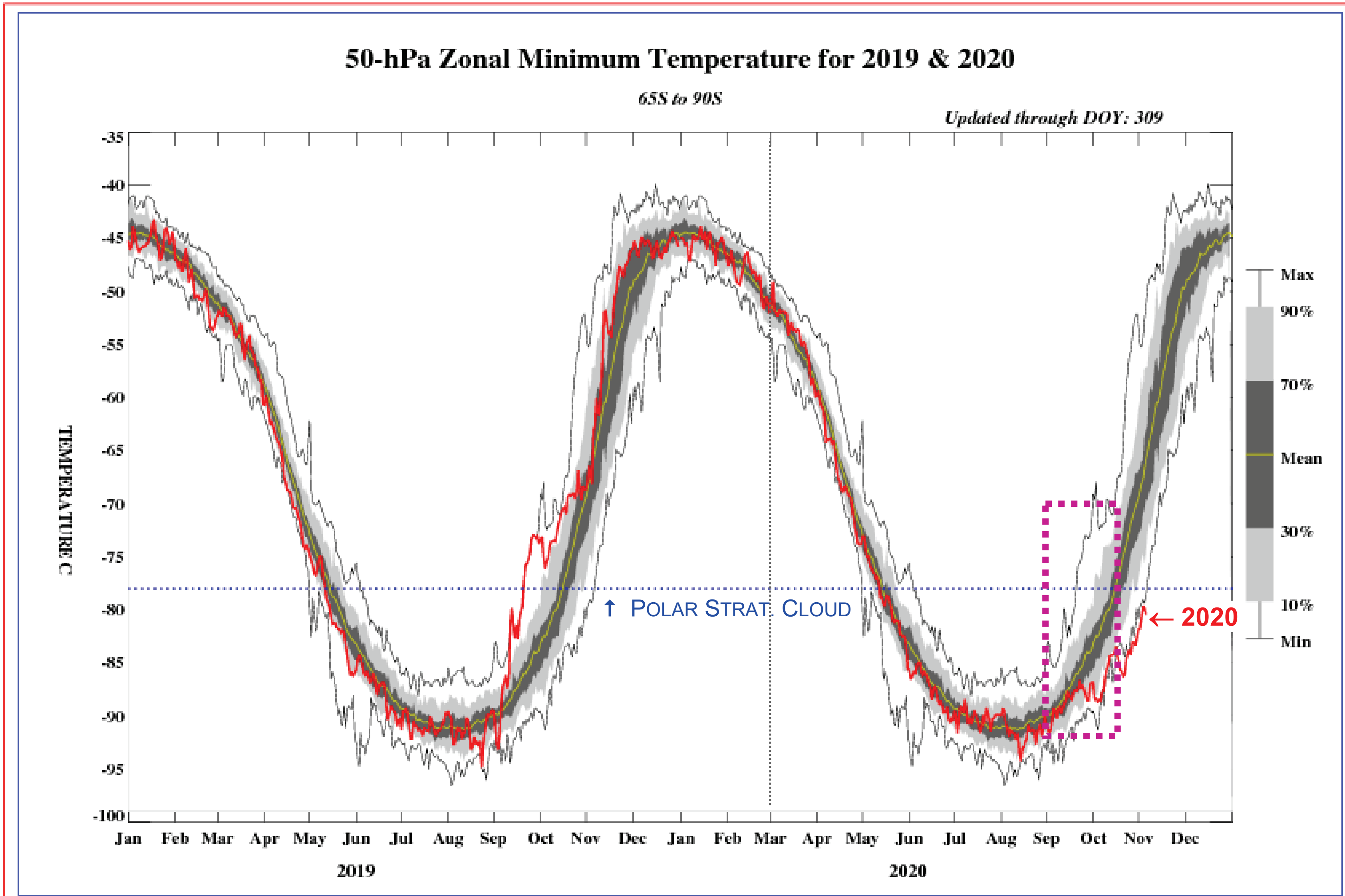
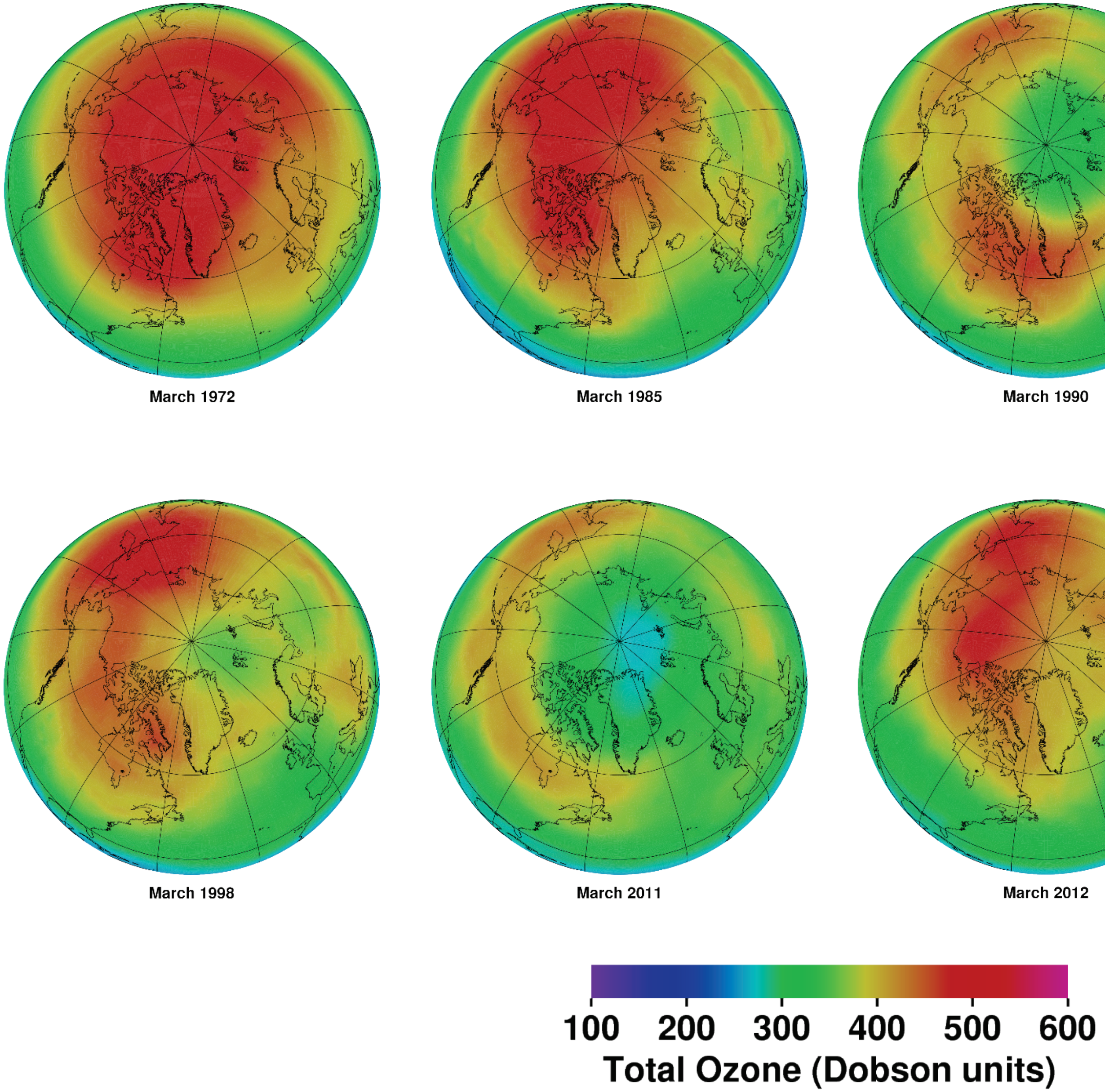
Antarctic Vortex Minimum Temperature: 2019-2020



https://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/50mbshlo.png

Arctic Ozone versus Time



Polar Ozone Depletion: NH and SH

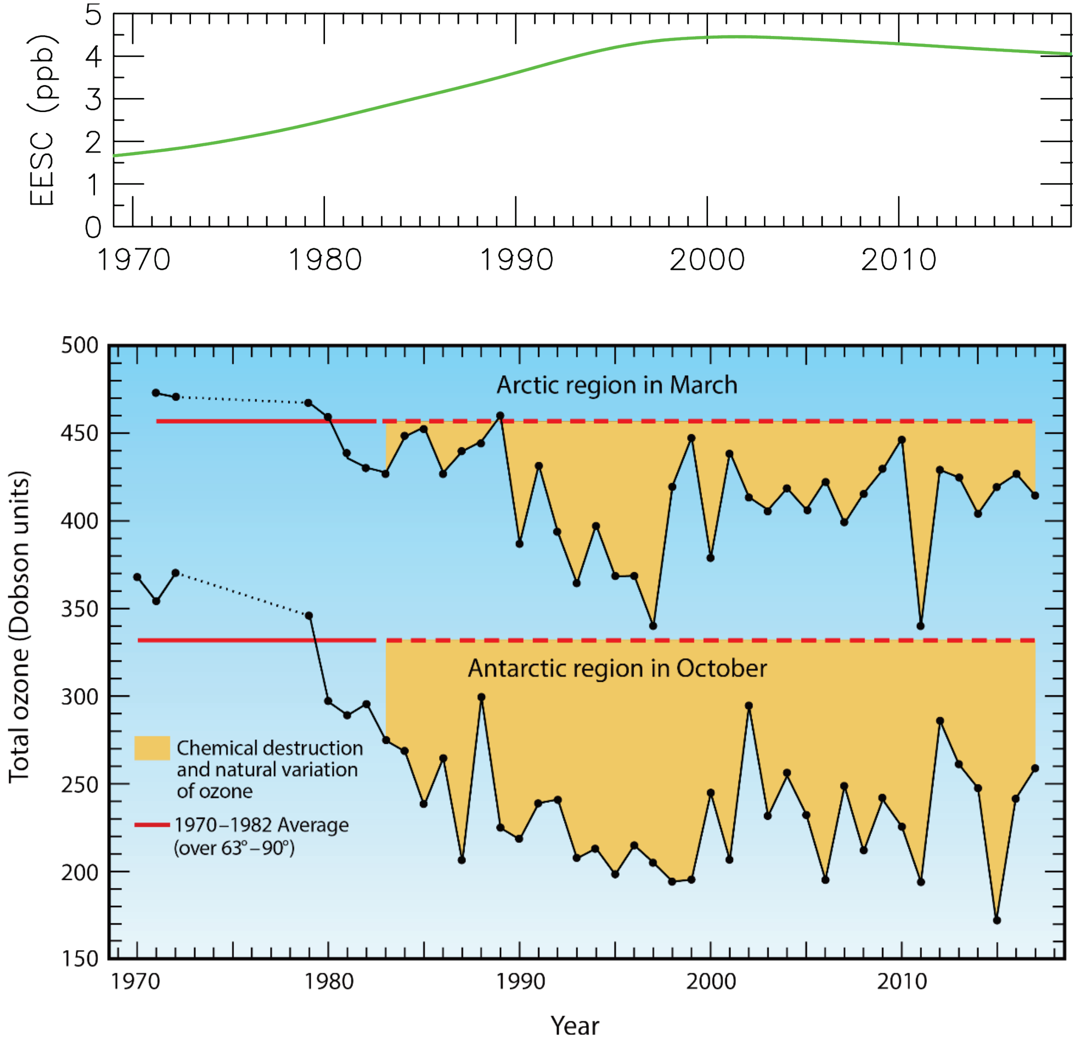
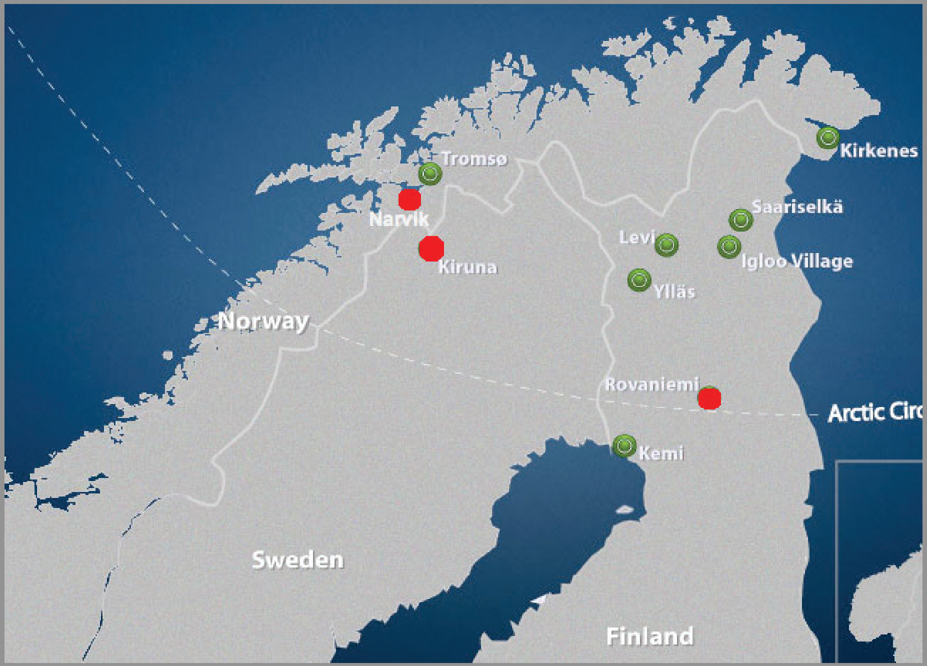


Fig Q11-1, *WMO/UNEP Twenty QAs Ozone*

Arena Arctica, Kiruna, Sweden

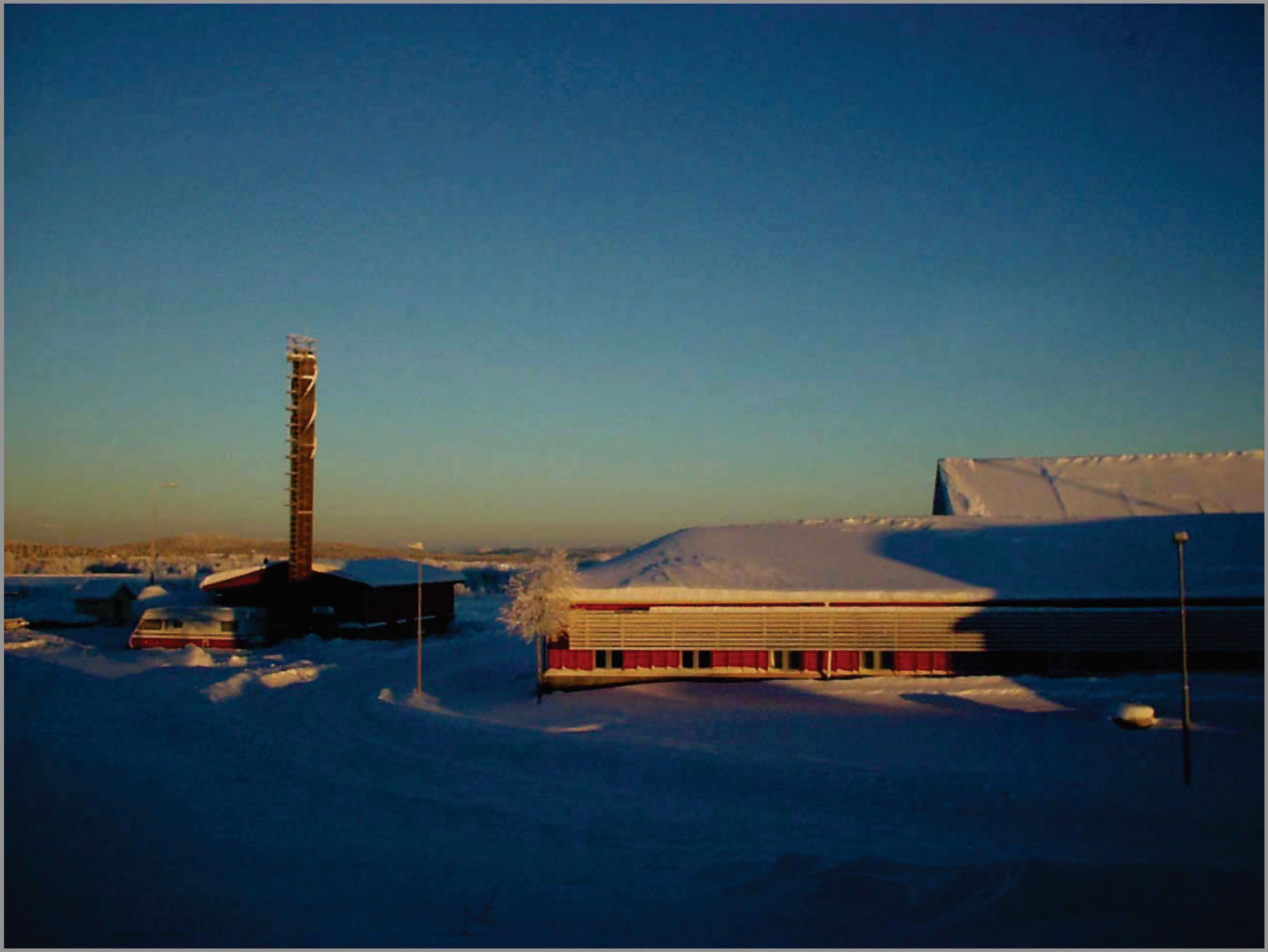


https://thiswendycity.wordpress.com/magic-laplands/

Arena Arctica, Kiruna, Sweden

Ross Salawitch (JPL), Markus Rex (Alfred Wegener Institute, Potsdam), and Rolf Mueller (FZ Juelich Research Center) are part of the data analysis and flight planning team. Here, they appear to be having too much fun for either data analysis or flight planning.

Arena Arctica, Kiruna, Sweden



View from Markus, Ross, and Rolf's office on an unusual day (bright, sunny, clear skies).

Kiruna, Sweden

Kiruna, Sweden

Kiruna, Sweden

Kiruna, Sweden

Kiruna, Sweden

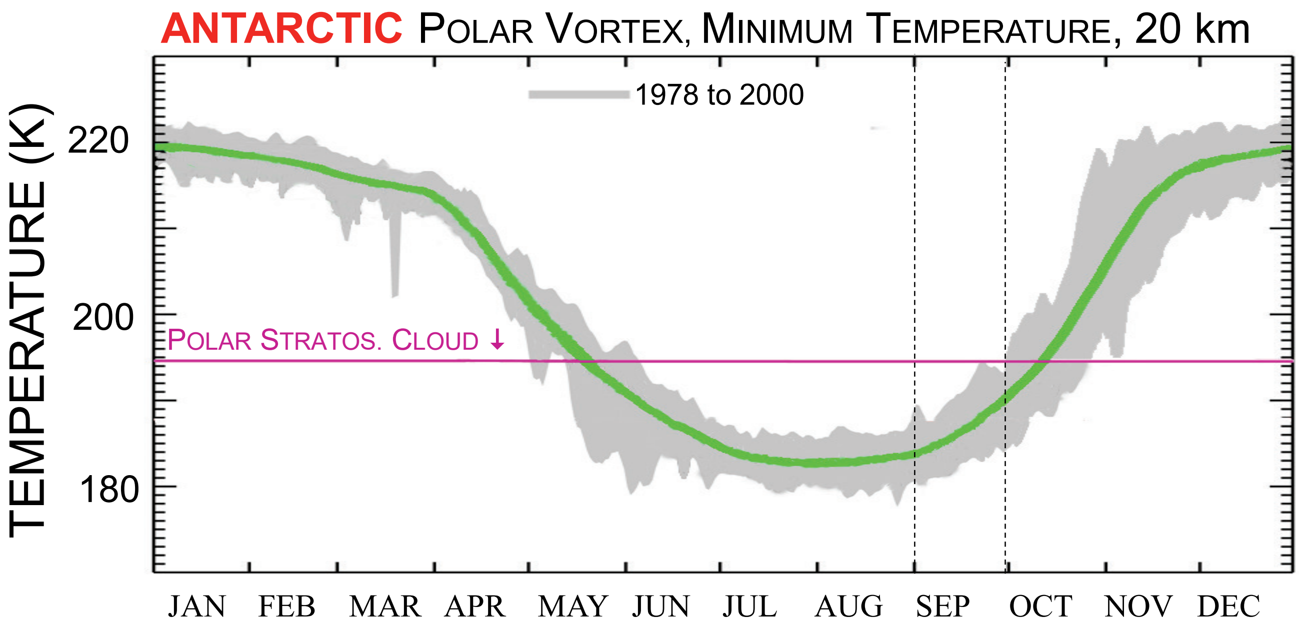
Arctic Overview

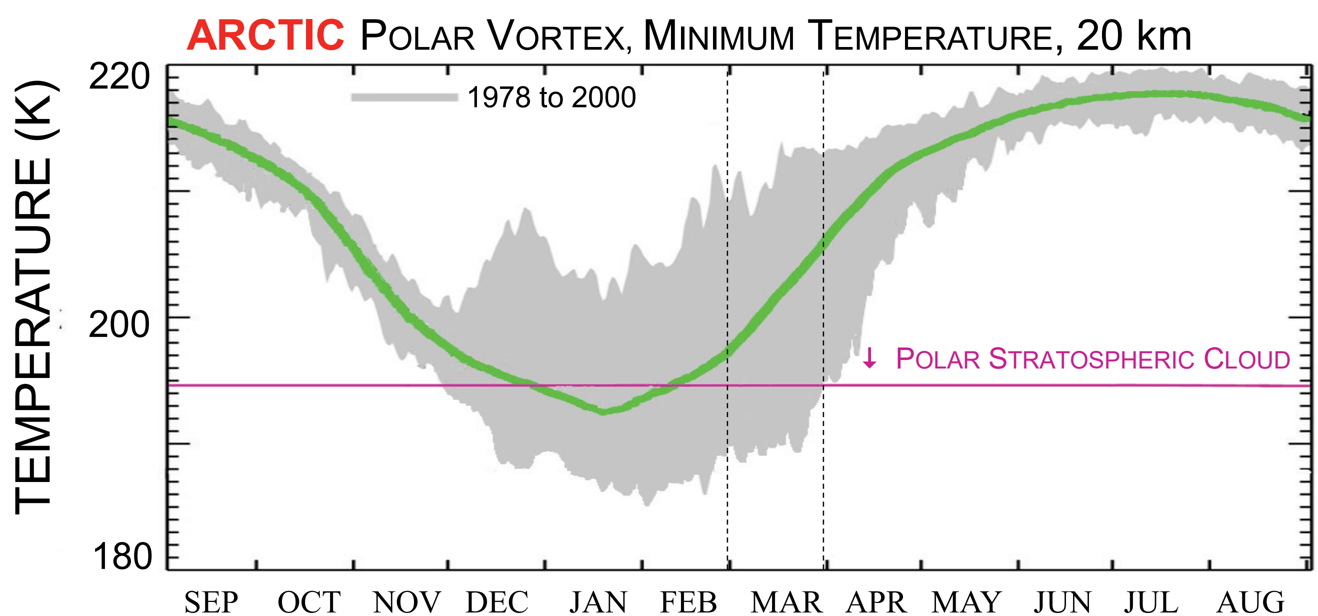
Arctic vortex (polar stratosphere):

- Always warmer than typical Antarctic winter
- Tremendous year to year variability in temperature
- Chemical ozone loss occurs *only* during cold winters
- Enough HNO_3 usually remains so that ClO recovers to $ClNO_3$:
  faster ClO de-activation (less ozone depletion) compared to Antarctic

All of this is due to hemispheric differences in atmospheric dynamics:

- More vigorous circulation in NH due to much more land-sea contrast, which triggers poleward transport of heat by atmospheric motions
  (Antarctic ice sheet suppresses poleward transport of heat by atmosphere)
- Stronger circulation in NH leads to more disturbed vortex (warmer, less PSCs)





Data Courtesy P. Newman, NASA/GSFC

Minimum Temperature: NH and SH

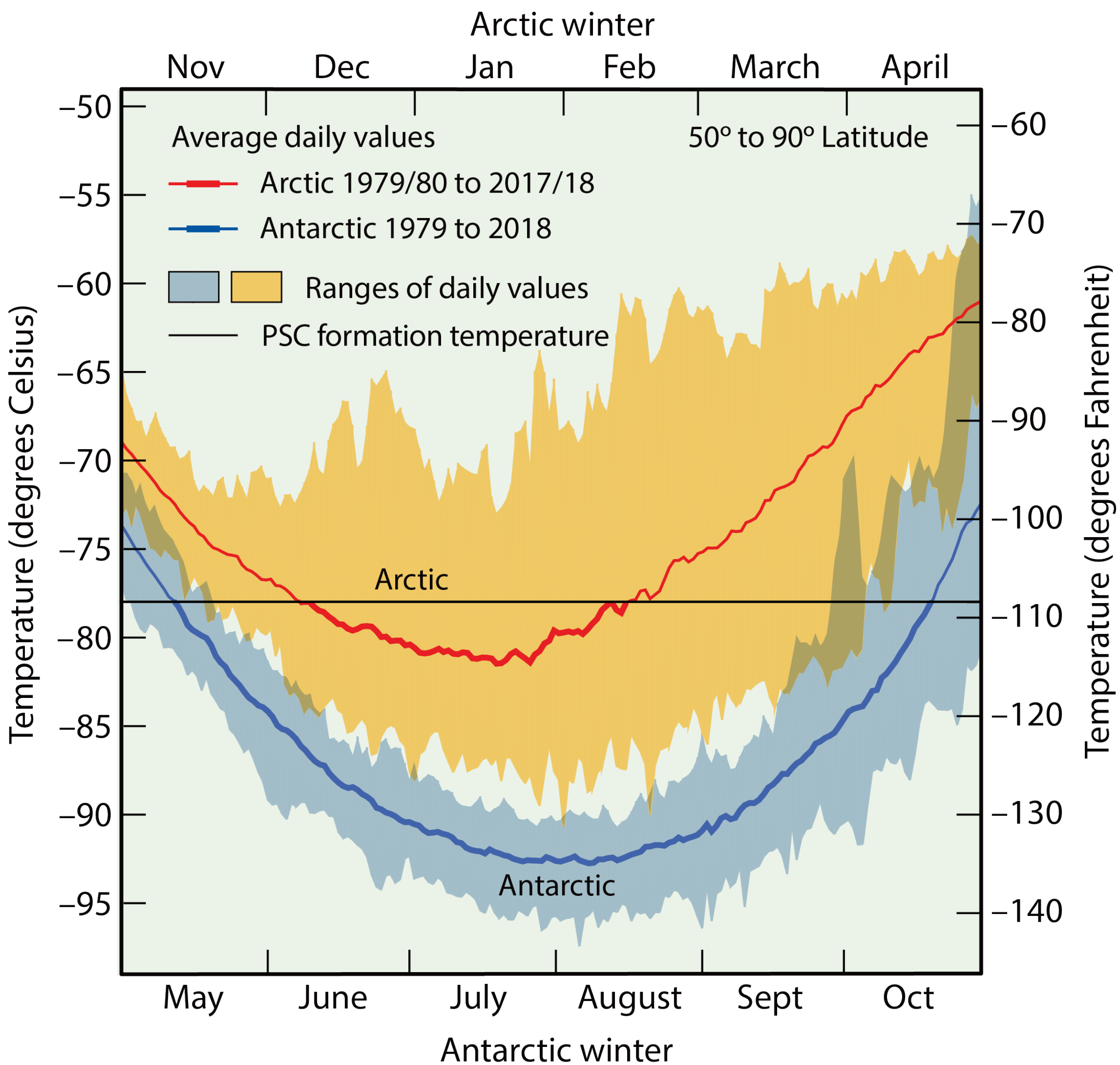


Fig Q9-1, *WMO/UNEP Twenty QAs Ozone*

The Stratosphere Cools as the Surface Warms !

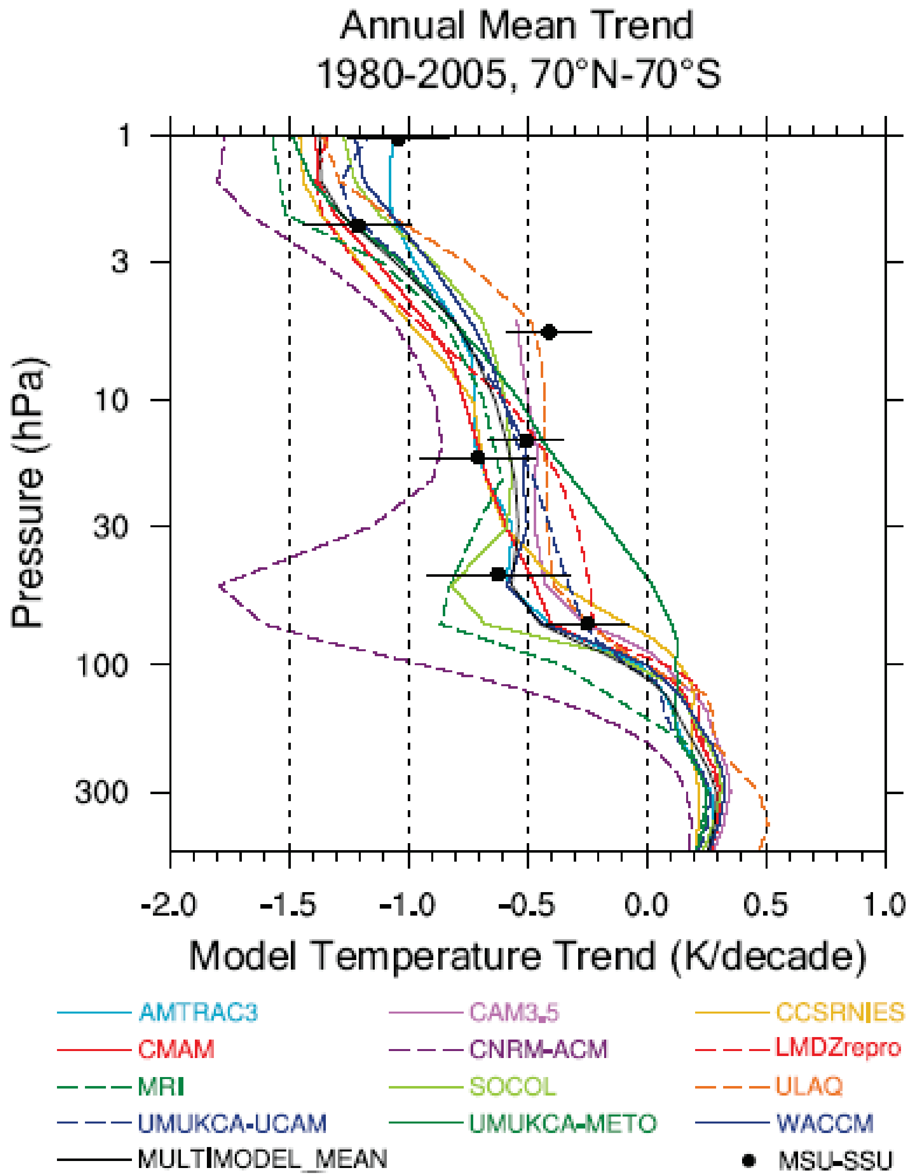


Figure 4−11, WMO/UNEP (2011)

The Stratosphere Cools as the Surface Warms !

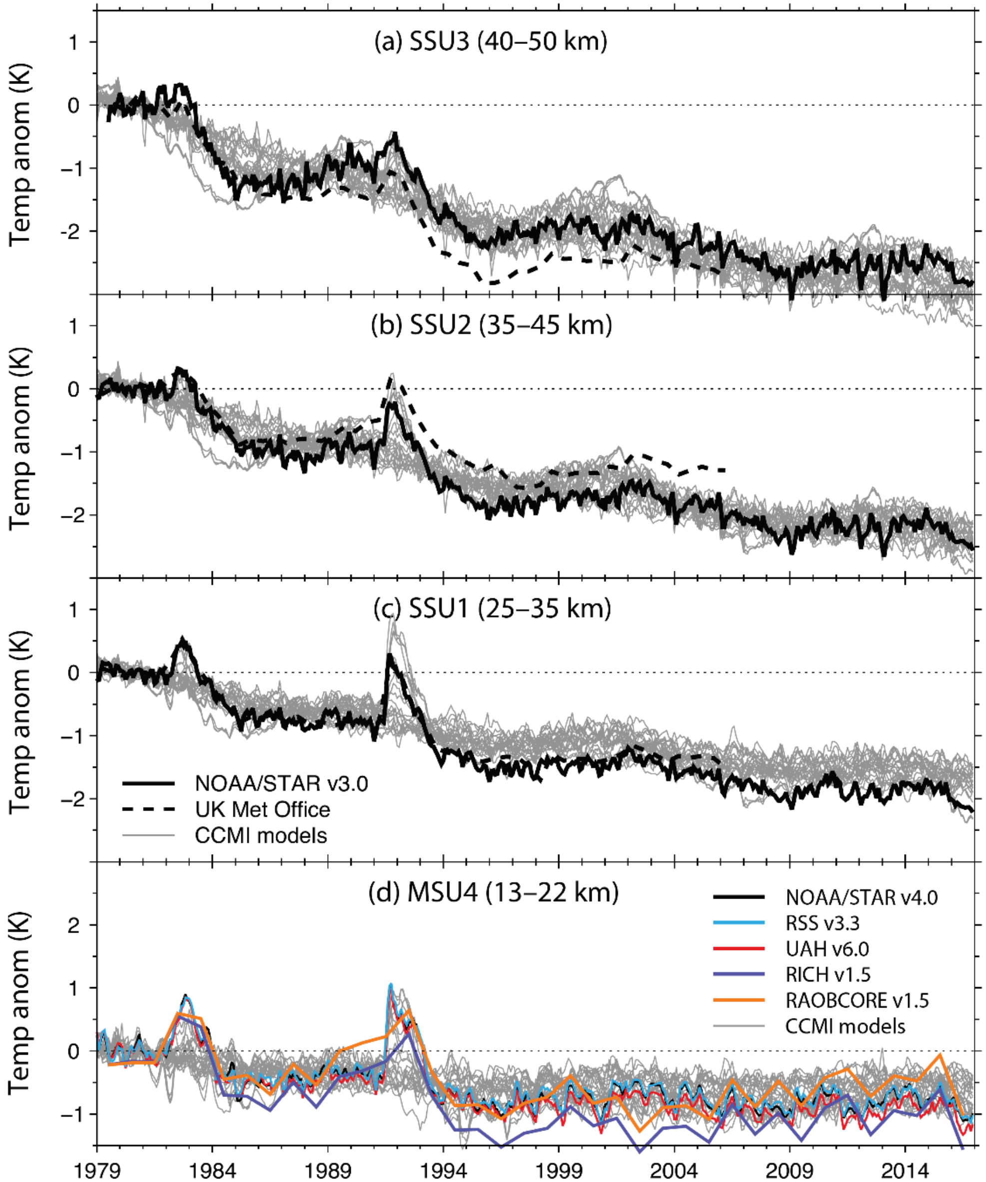
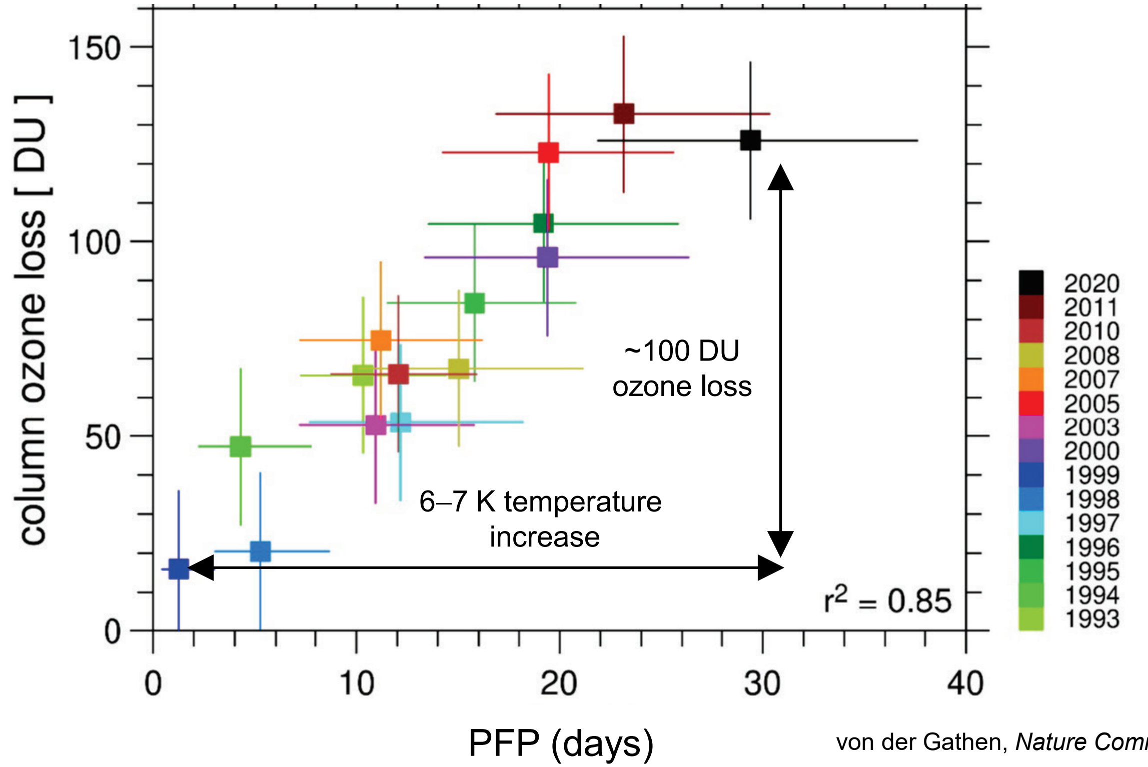


Figure 5-4. Time series of global mean stratospheric temperature anomalies from 1979 to 2016. Panels show SSU Channels 3, 2, 1 (SSU3, SSU2, SSU1; a, b, c) and MSU channel 4 (MSU4; d) for the altitude ranges, datasets, and model outputs indicated in the legends. Gray lines indicate results from a total of 23 ensemble members across 14 Chemistry-Climate Model Initiative (CCMI) models for the REF-C2 experiment, weighted by the appropriate satellite weighting function for comparison with observations. All data in panel d are shown as monthly averages except the UK Met Office dataset, which uses 6-month averages, and the two radiosonde datasets, which are annual means. The radiosonde data are as in Figure 2.8 of Blunden and Arndt (2017). Anomalies are shown relative to 1979–1981. Adapted from Maycock et al. (2018).

Figure 5-4, WMO/UNEP (2018)

Arctic Ozone Loss Varies as a function of PSC Formation Potential

Data:



von der Gathen, *Nature Communications*, submitted, 2020

- Surprisingly simple relationship between chemical loss of column ozone and volume of air exposed to PSC formation potential over winter, where

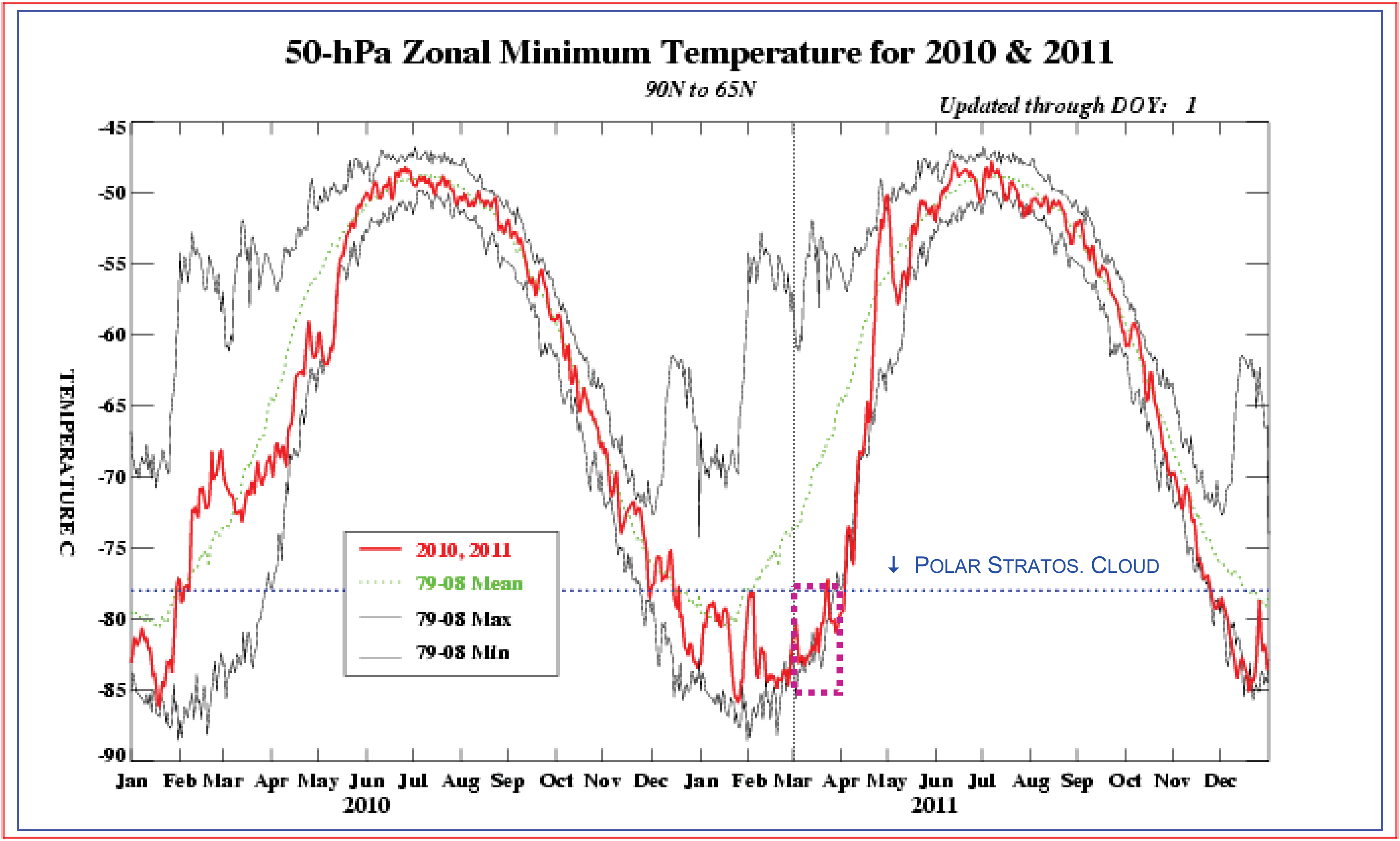
$$PFP = \int_{16\ \mathrm{Nov}}^{17\ \mathrm{Apr}} \frac{V_{PSC}(t)}{V_{VORTEX}(t)}\, dt$$

PFP is PSC Formation Potential

and V_{PSC} is the volume of the vortex where T is cold enough to allow for formation of PSCs, and V_{VORTEX} is the volume of the Arctic vortex

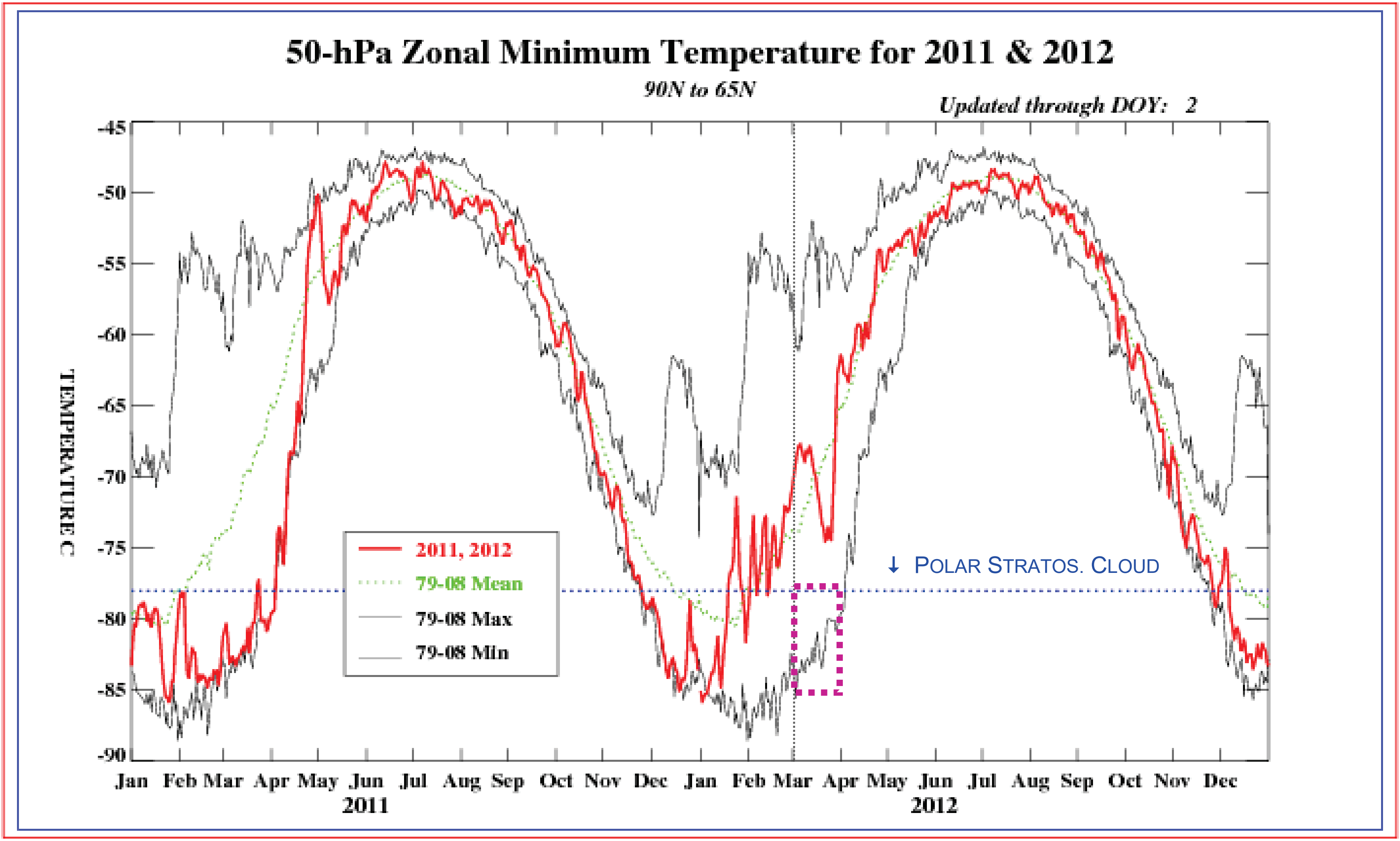
- Relation leads to estimate of ~15 DU additional loss of ozone per degree Kelvin cooling of *Arctic* stratosphere

Arctic Temperature: Mar 2011



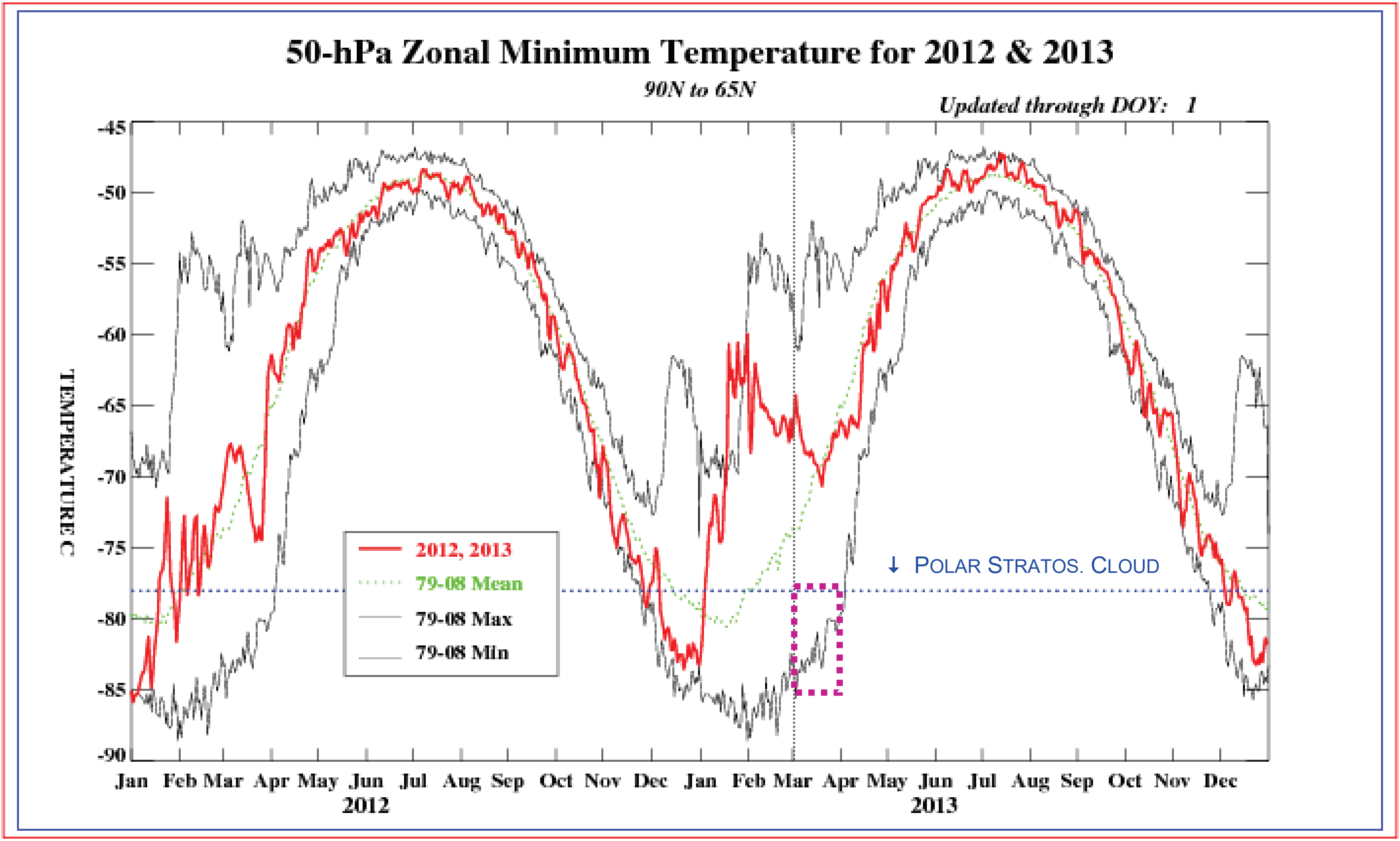
http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/archive/50mbnhlo_2011.gif

Arctic Temperature: Mar 2012



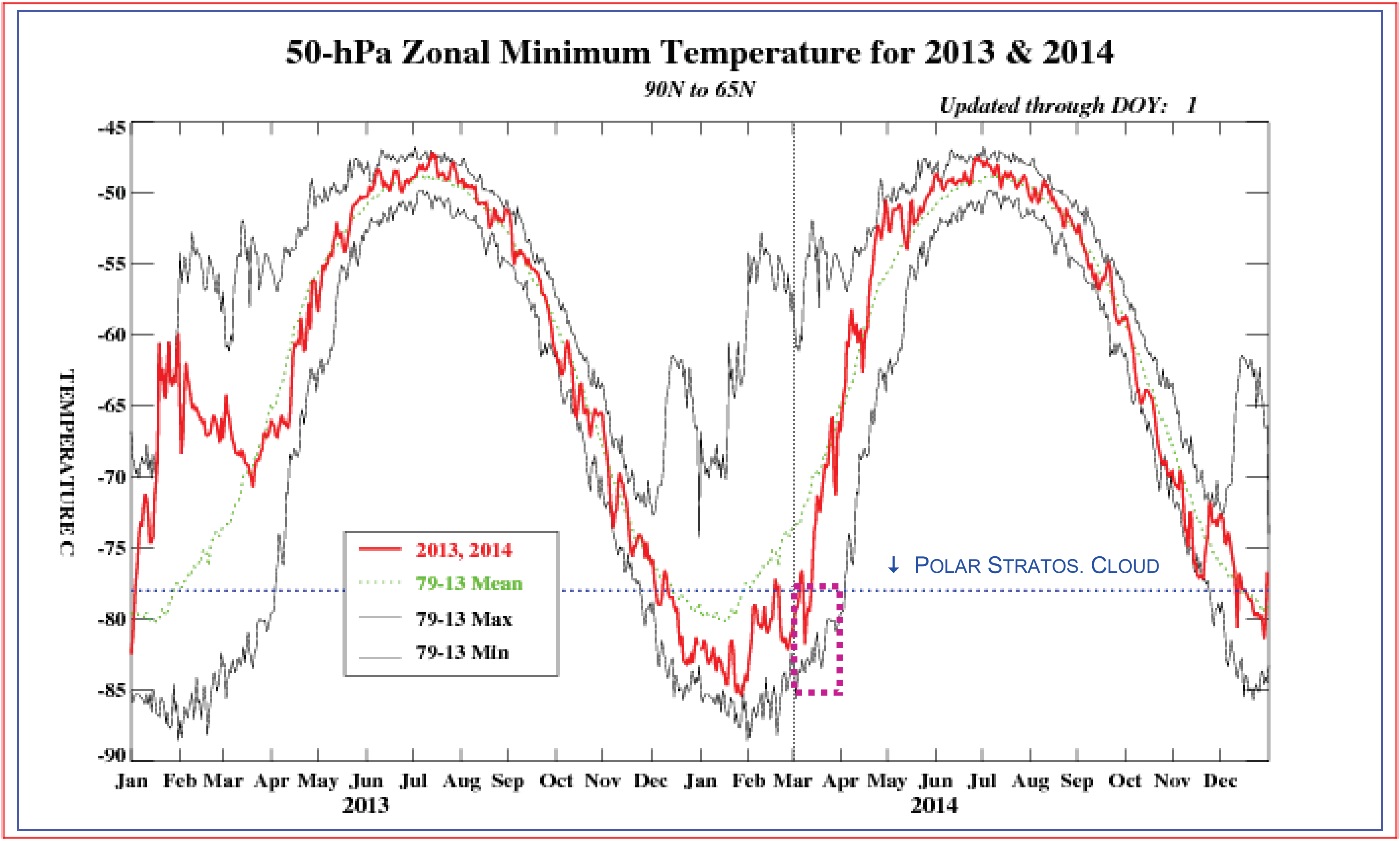
http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/archive/50mbnhlo_2012.gif

Arctic Temperature: Mar 2013



http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/archive/50mbnhlo_2013.gif

Arctic Temperature: Mar 2014



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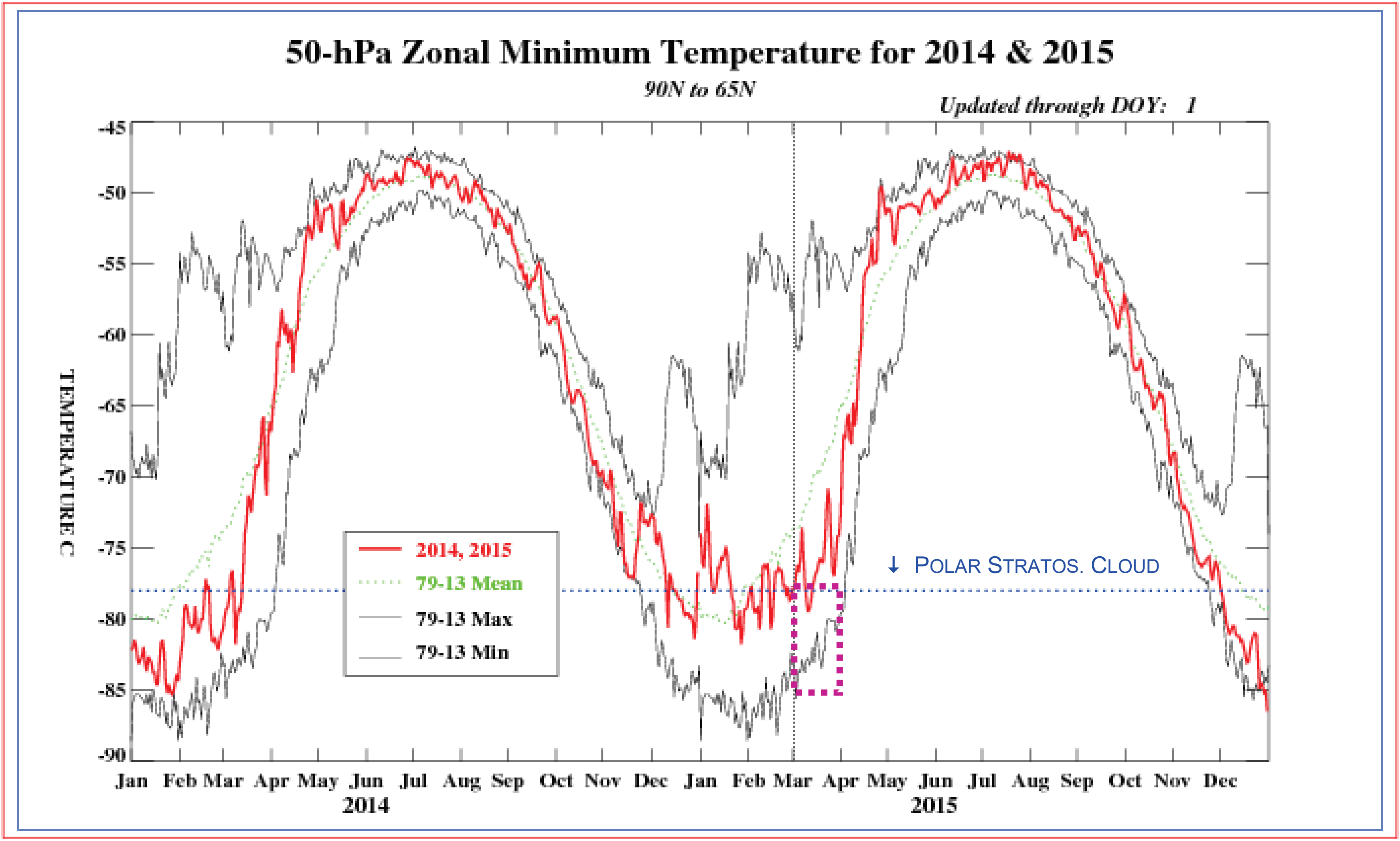
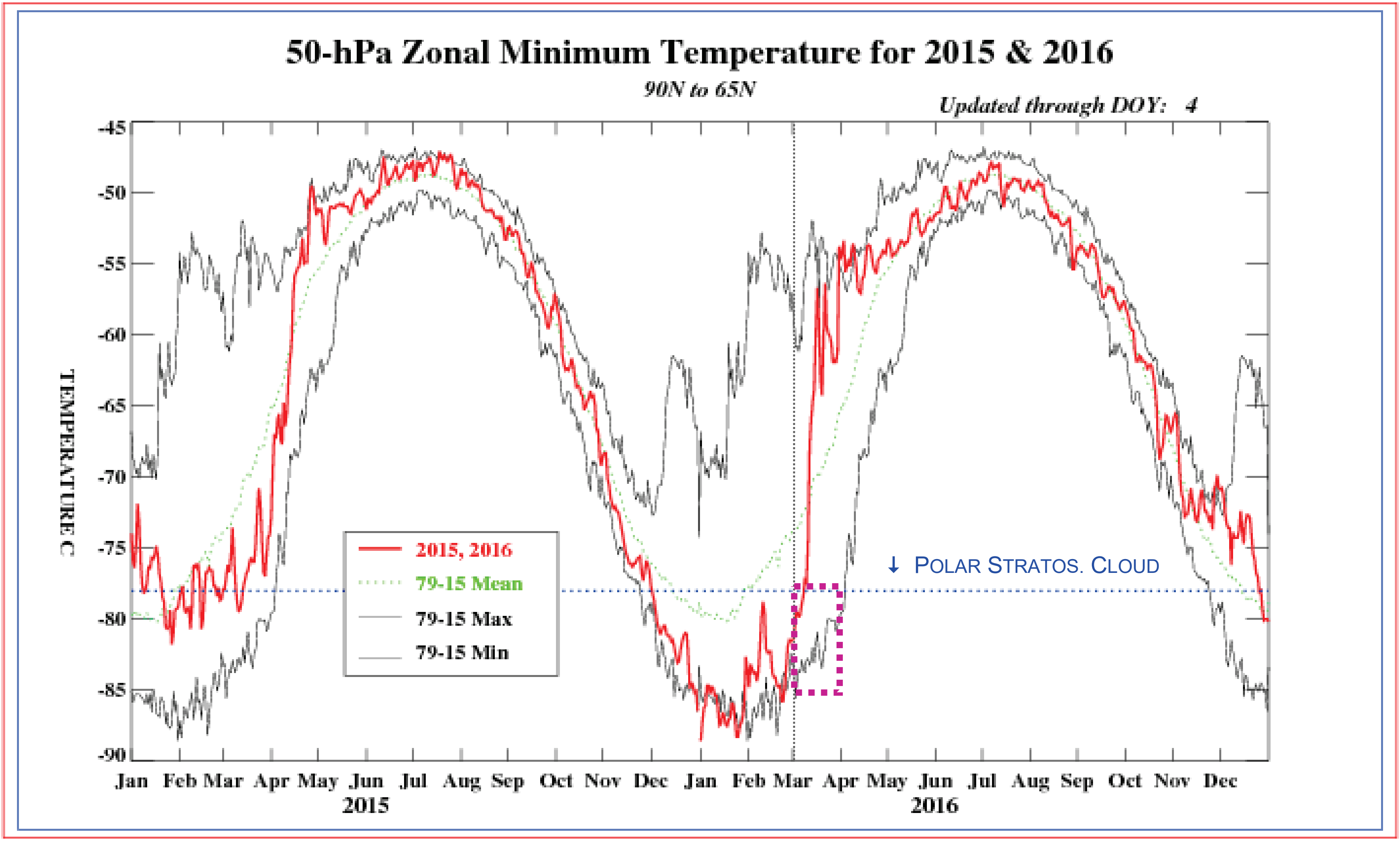
Arctic Temperature: Mar 2015



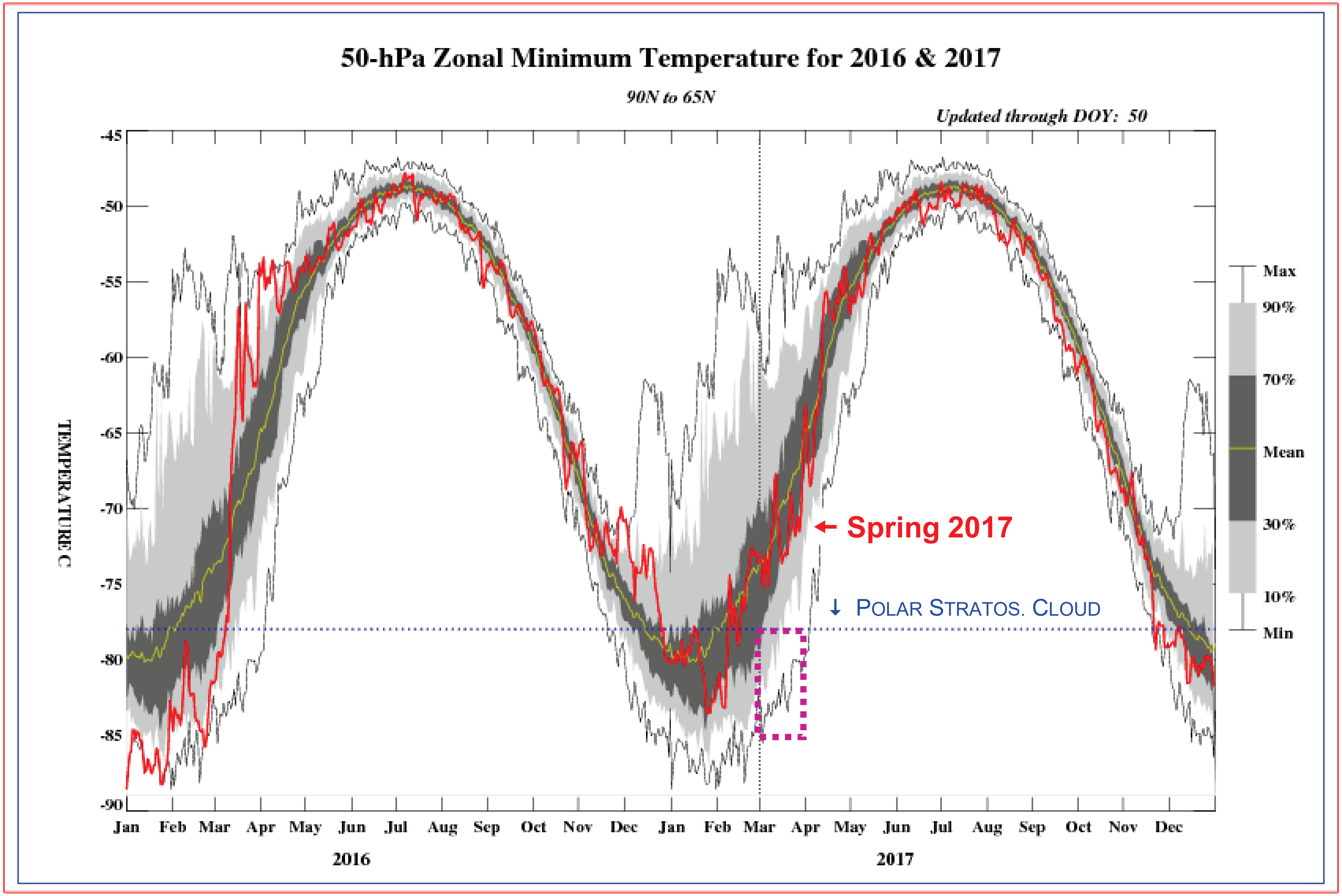
http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/archive/50mbnhlo_2015.gif

Arctic Temperature: Mar 2016



http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/archive/50mbnhlo_2016.gif

Arctic Temperature: Mar 2017



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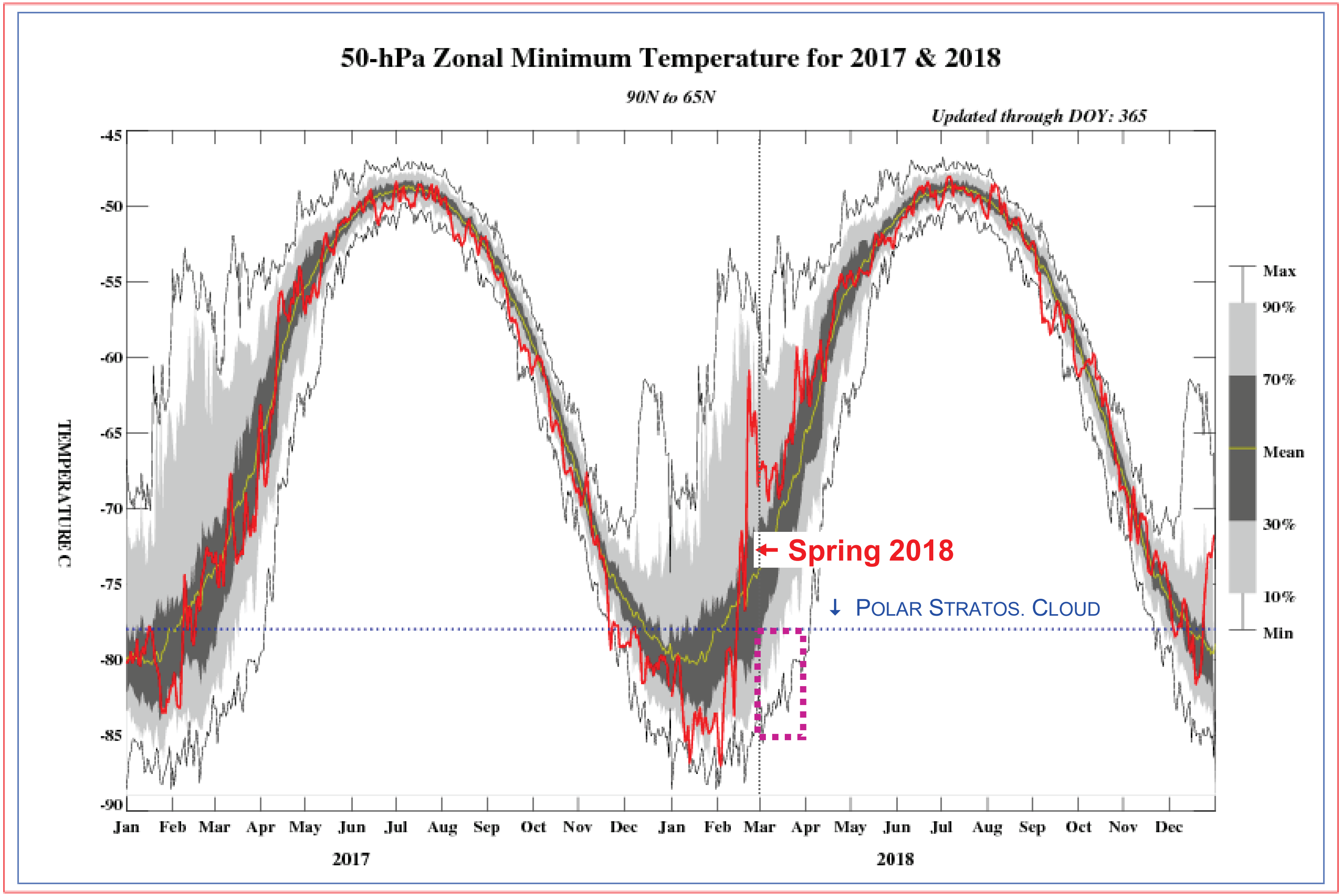
Arctic Temperature: Mar 2018



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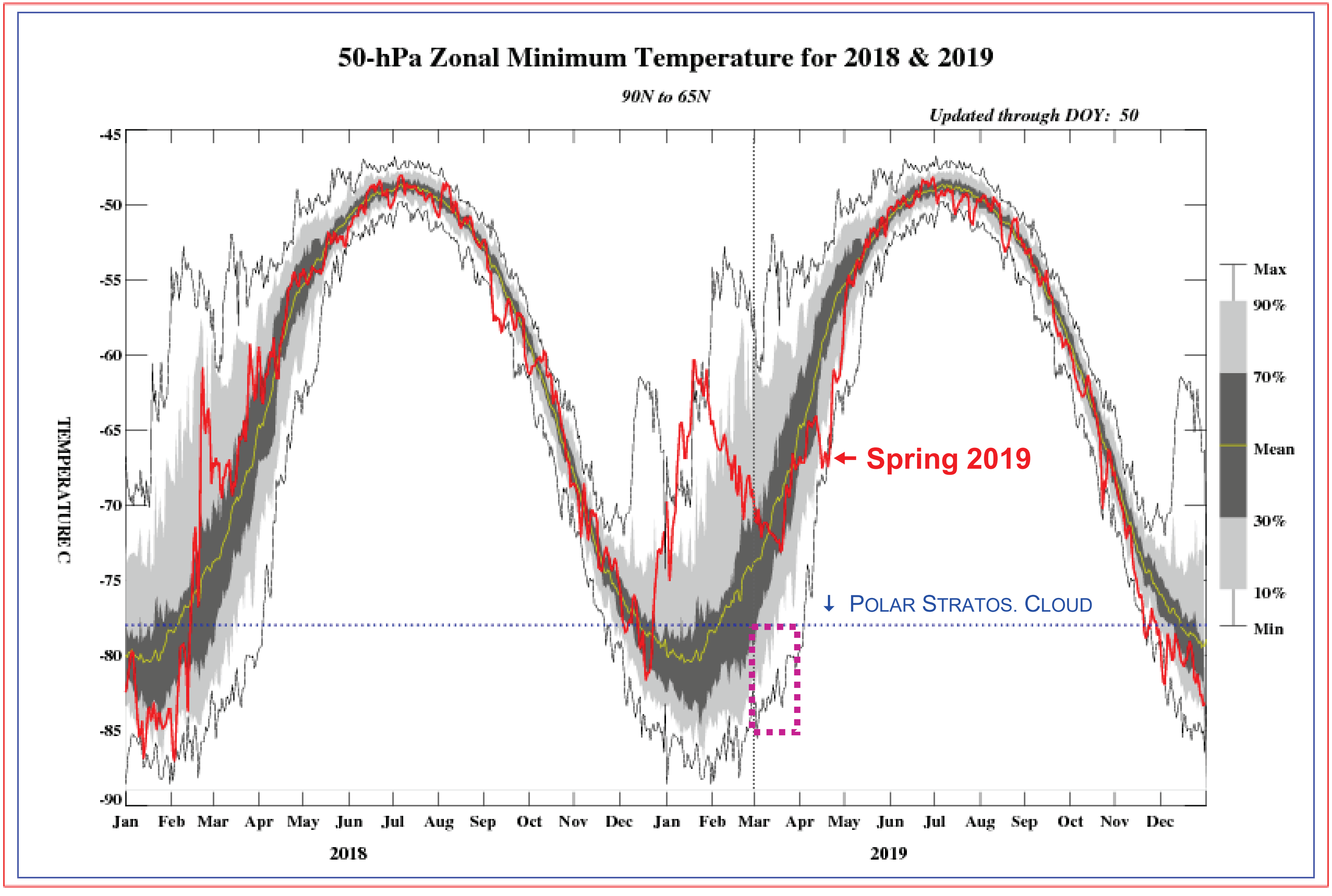
Arctic Temperature: Mar 2019



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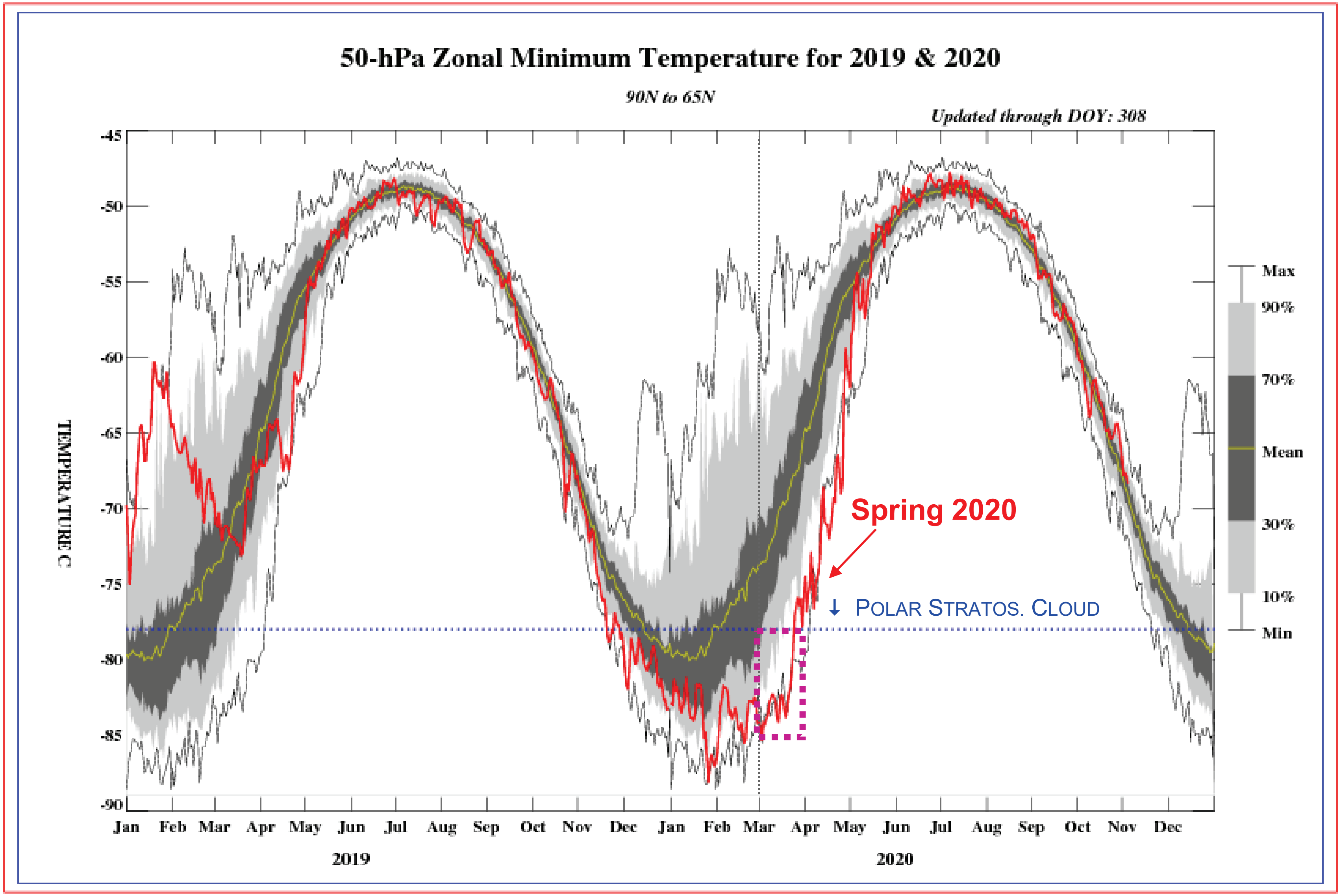
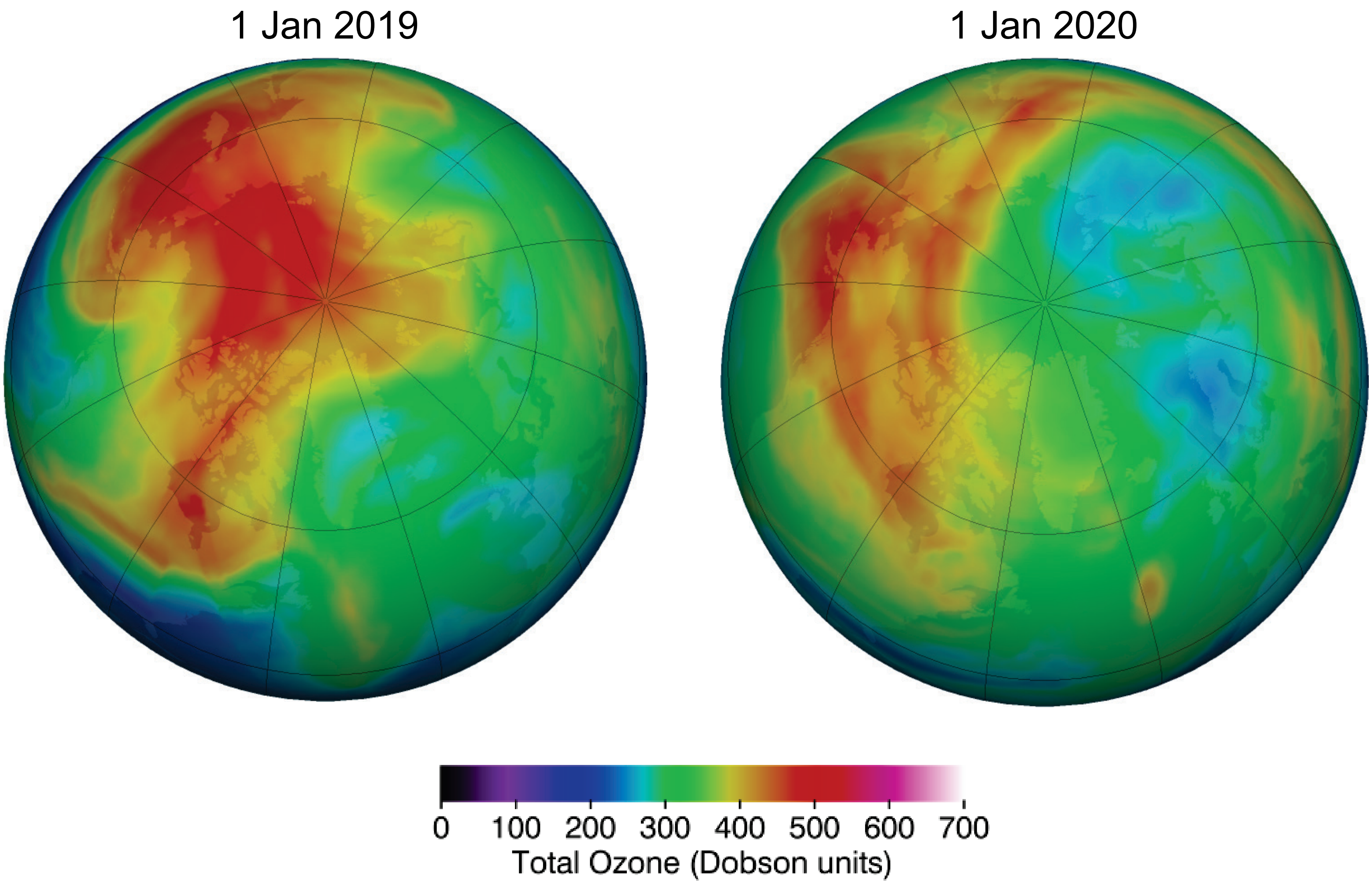
Arctic Temperature: Mar 2020



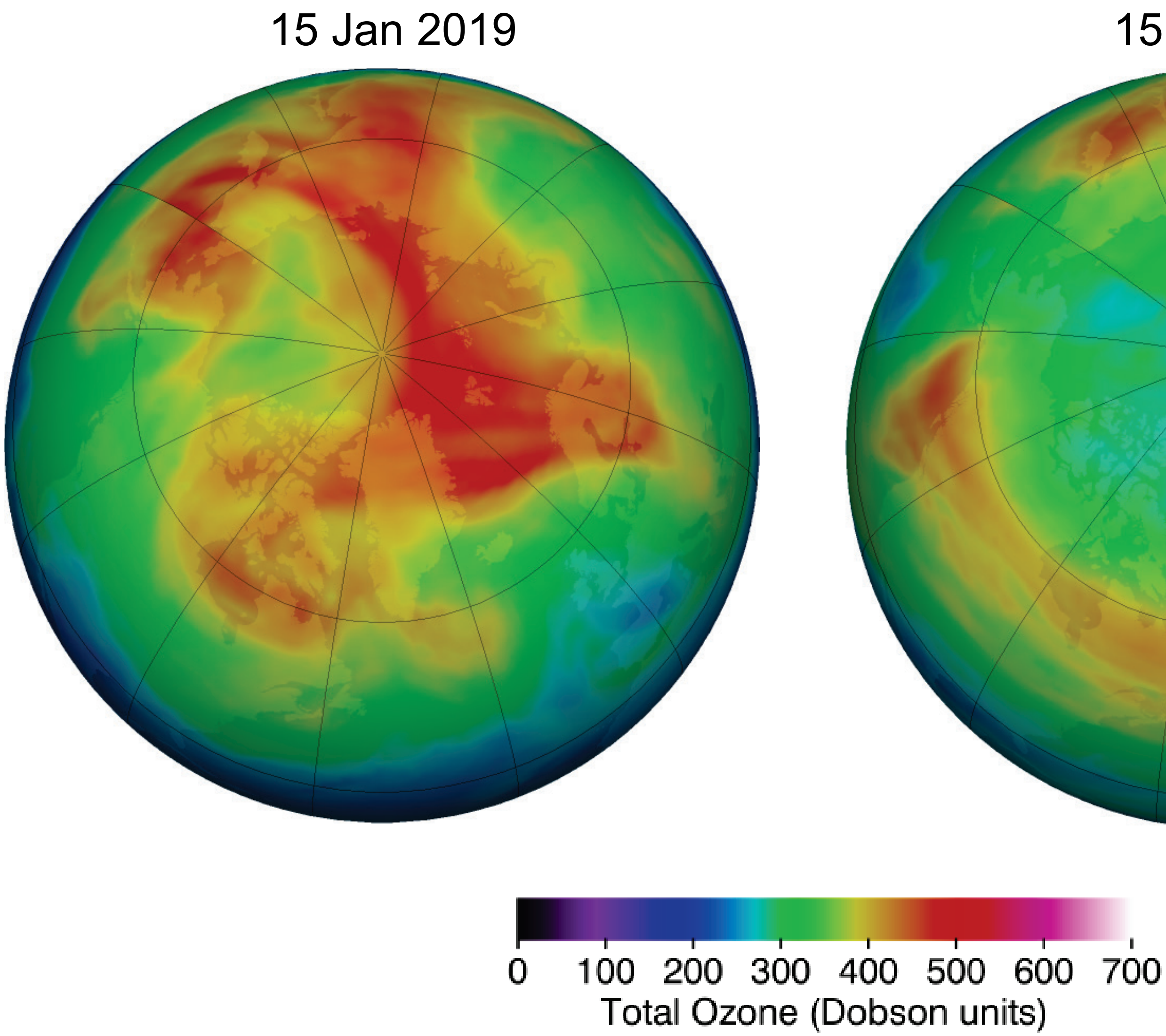
http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/50mbnhlo.png

Arctic Ozone: 2019 and 2020



https://ozonewatch.gsfc.nasa.gov/monthly/monthly_2019-04_NH.html

Arctic Ozone: 2019 and 2020



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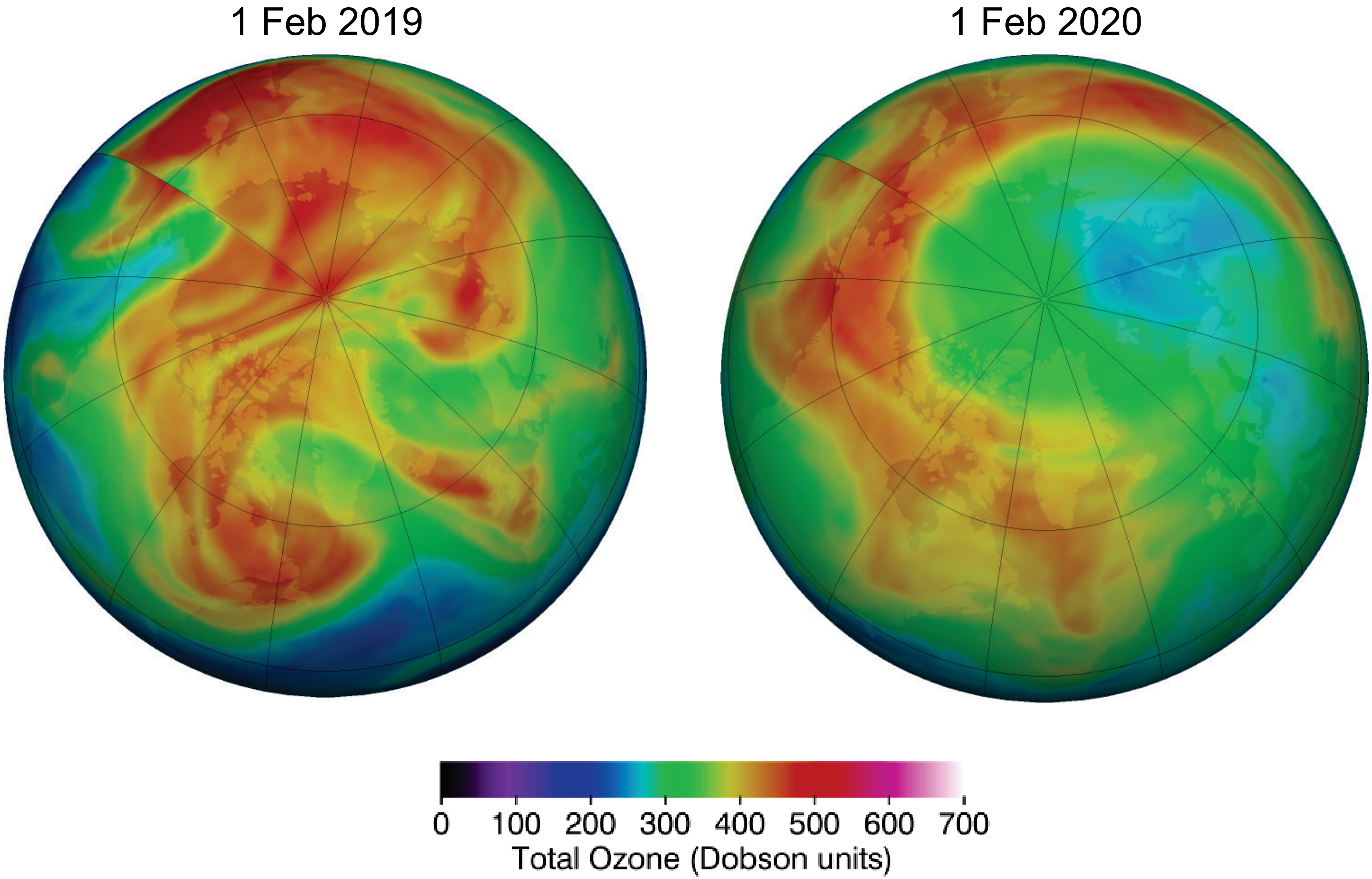

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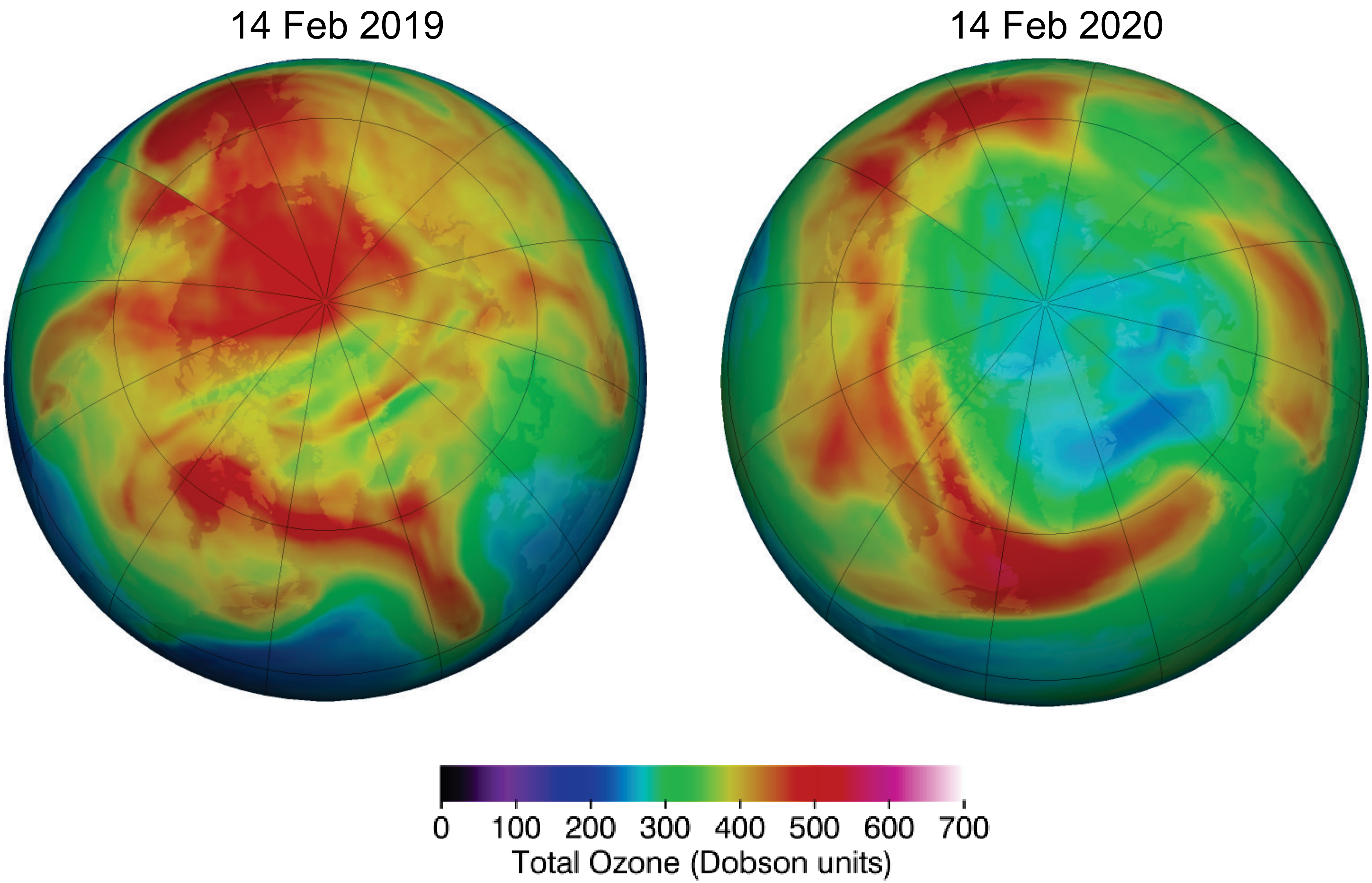

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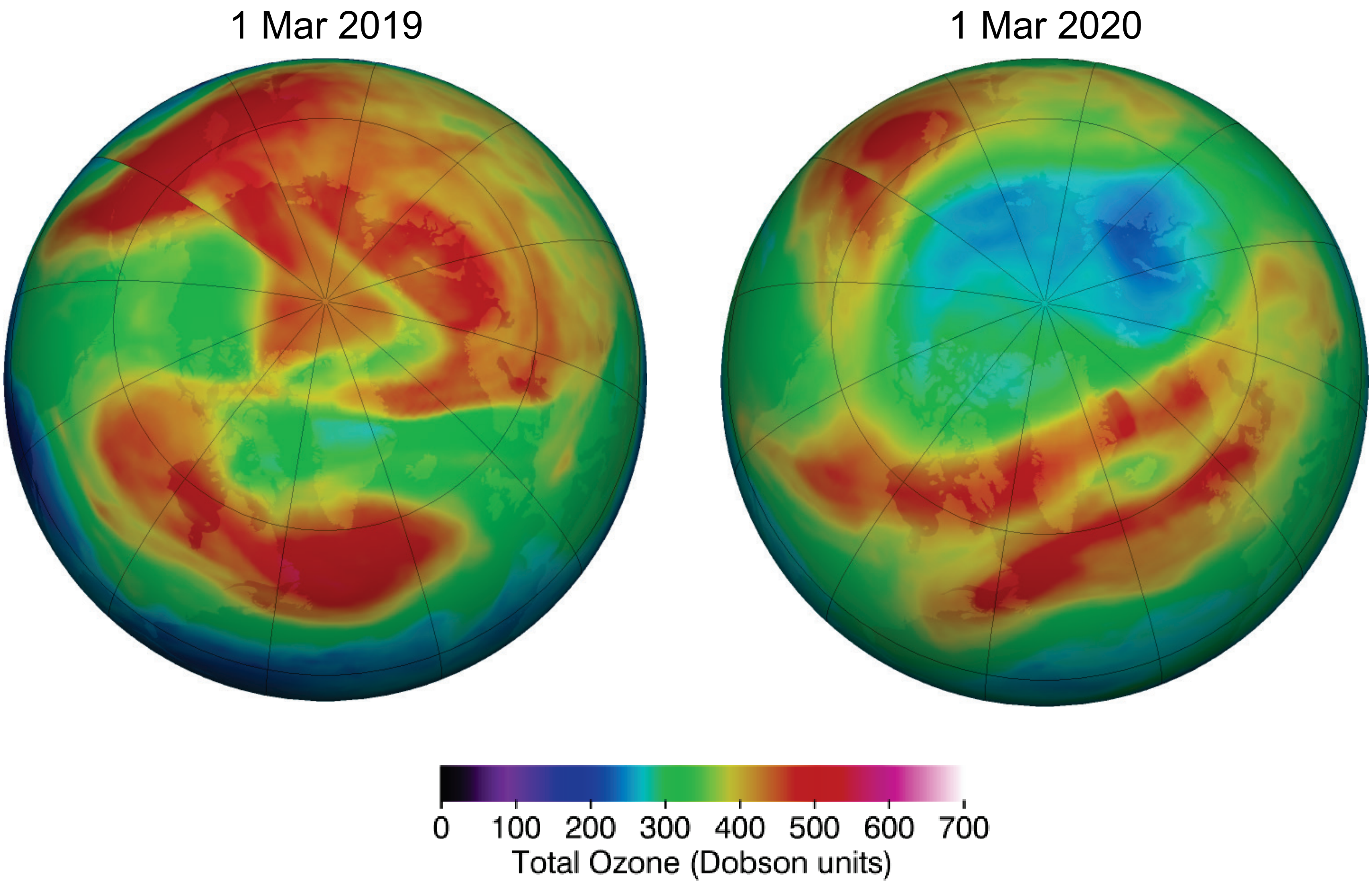

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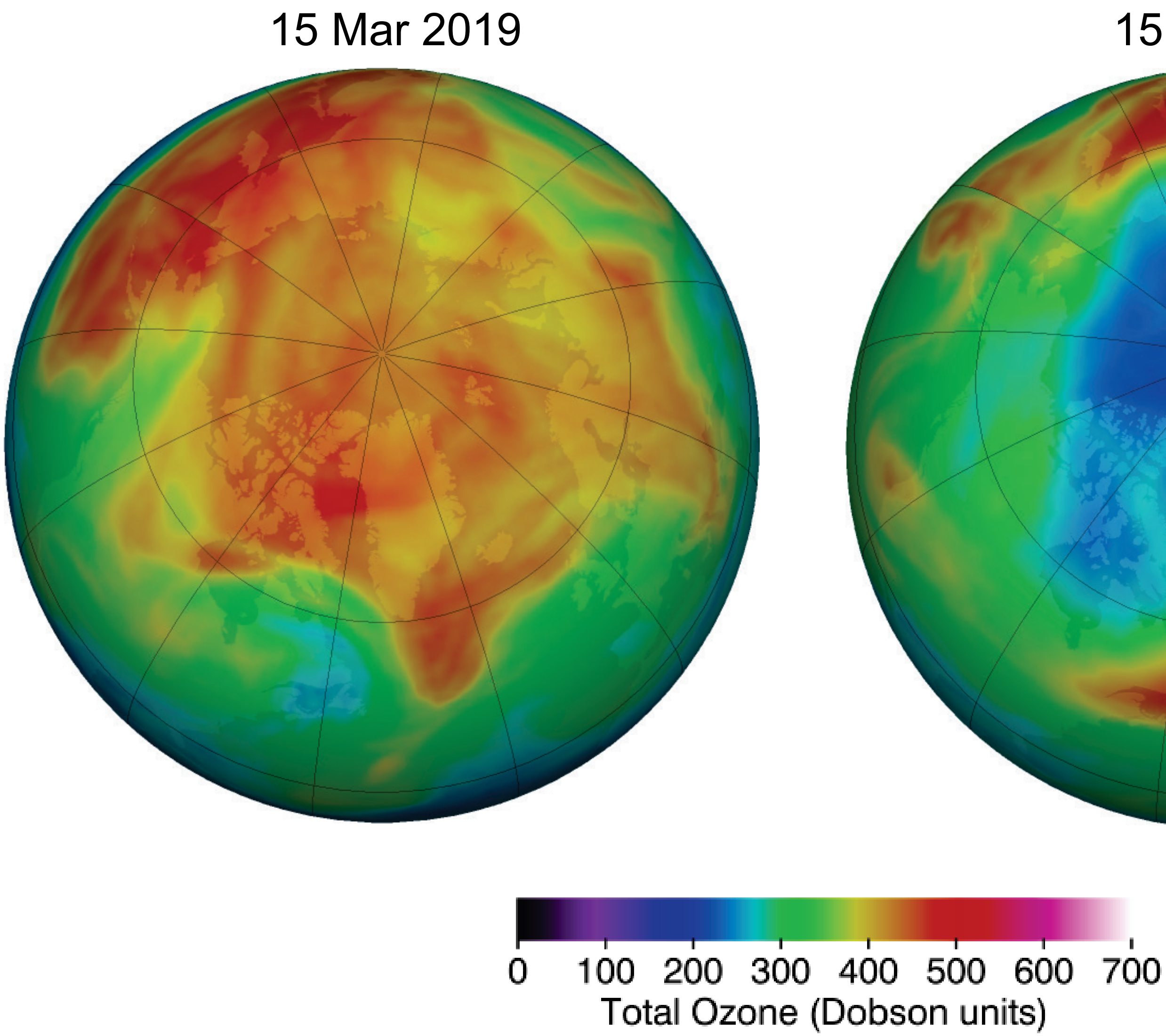
Arctic Ozone: 2019 and 2020



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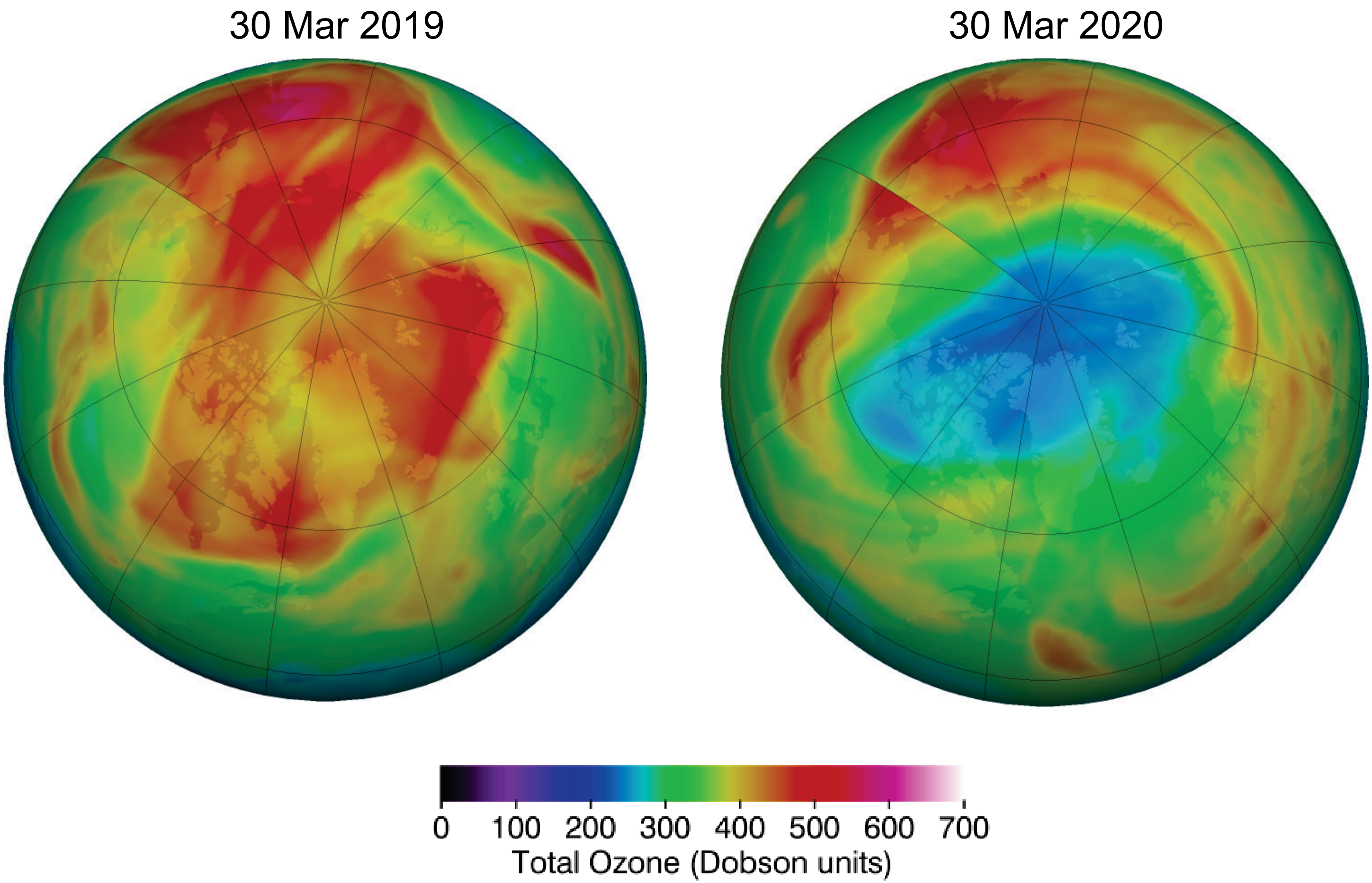
Arctic Ozone: 2019 and 2020



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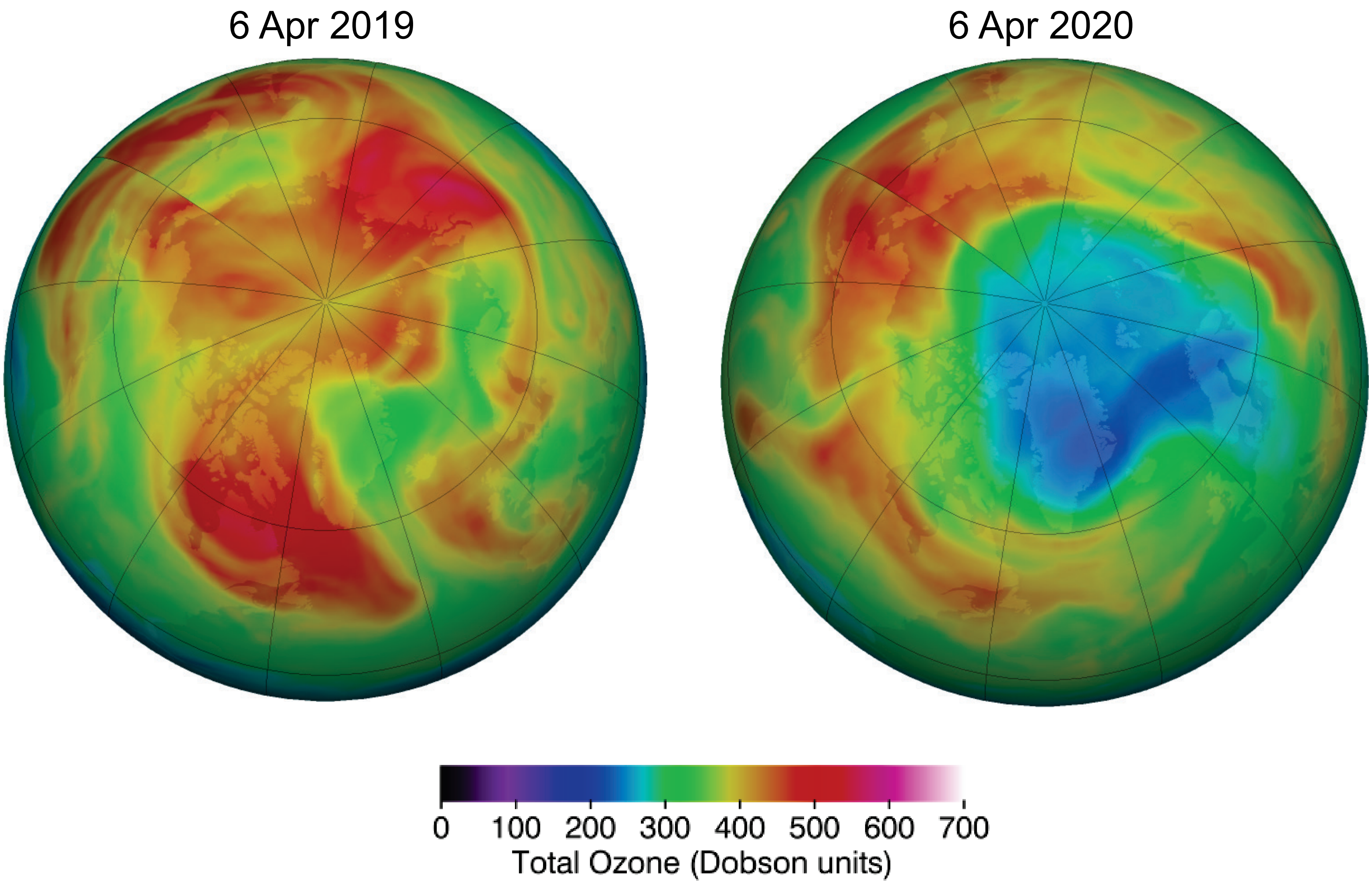
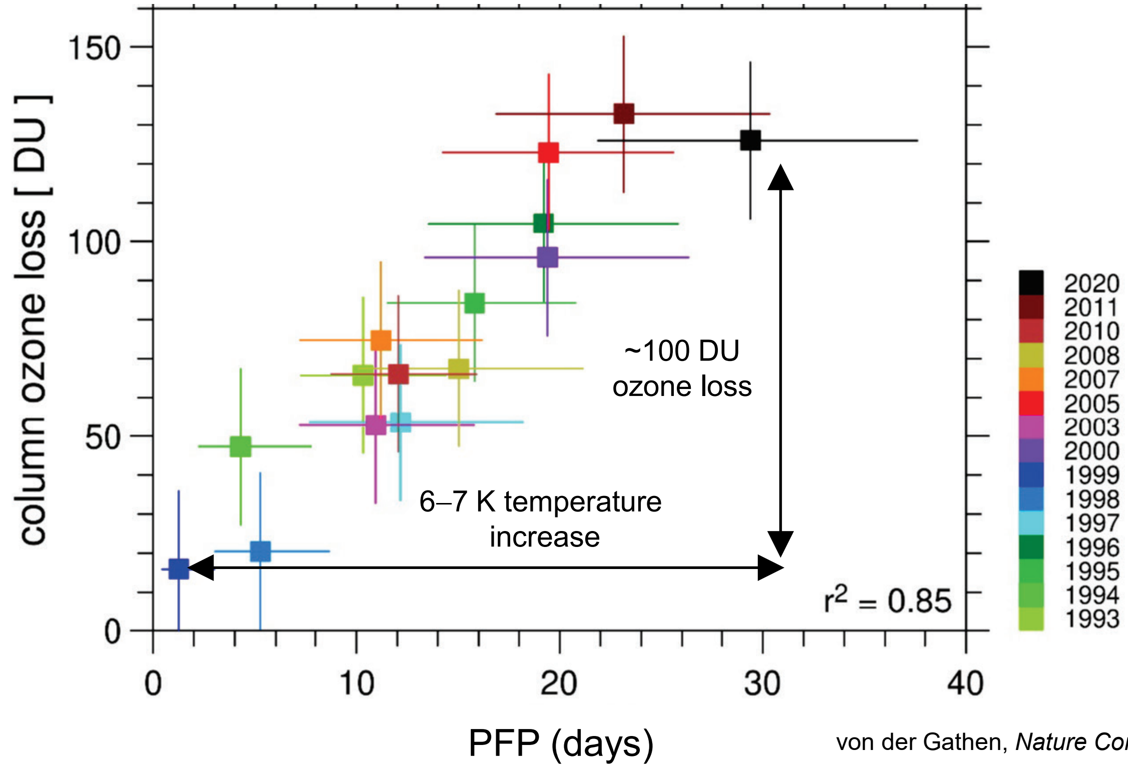


http://www.cpc.ncep.noaa.gov/products/stratosphere/temperature/50mbnhlo.png

Arctic Ozone Loss Varies as a function of PSC Formation Potential

Data:



von der Gathen, *Nature Communications*, submitted, 2020

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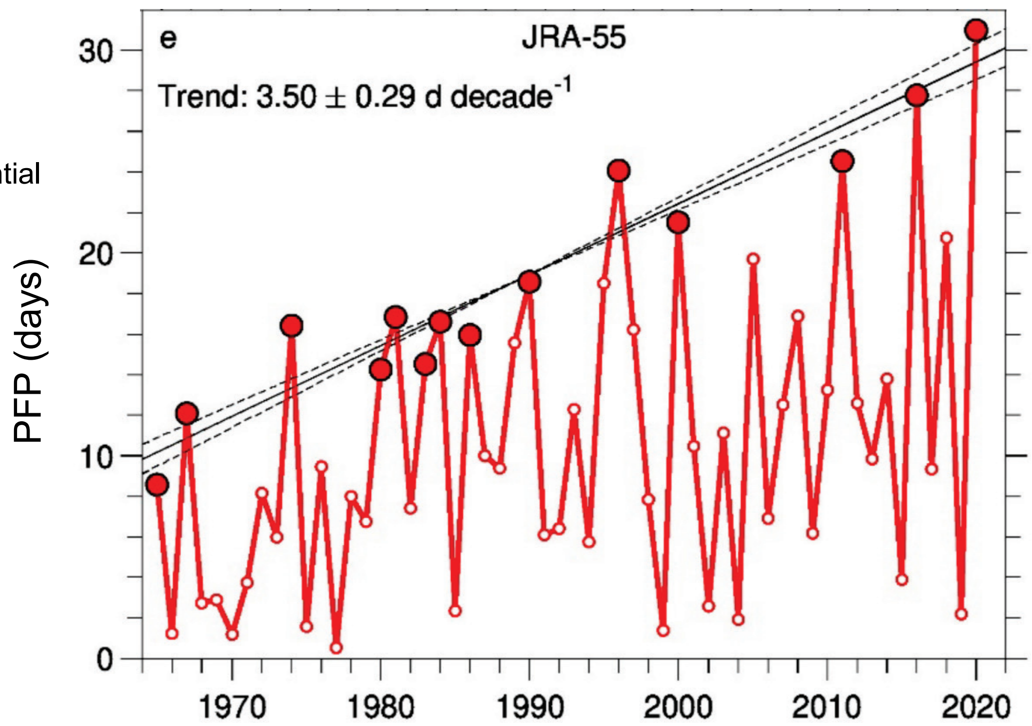
- Relation leads to estimate of ~15 DU additional loss of ozone per degree Kelvin cooling of *Arctic* stratosphere

Cold Arctic Winters Tend to Exhibit Larger PFP as a Function of Time

More Data:

PFP is PSC Formation Potential



von der Gathen, *Nature Communications*, submitted, 2020

PSC Formation Potential in Arctic Vortex

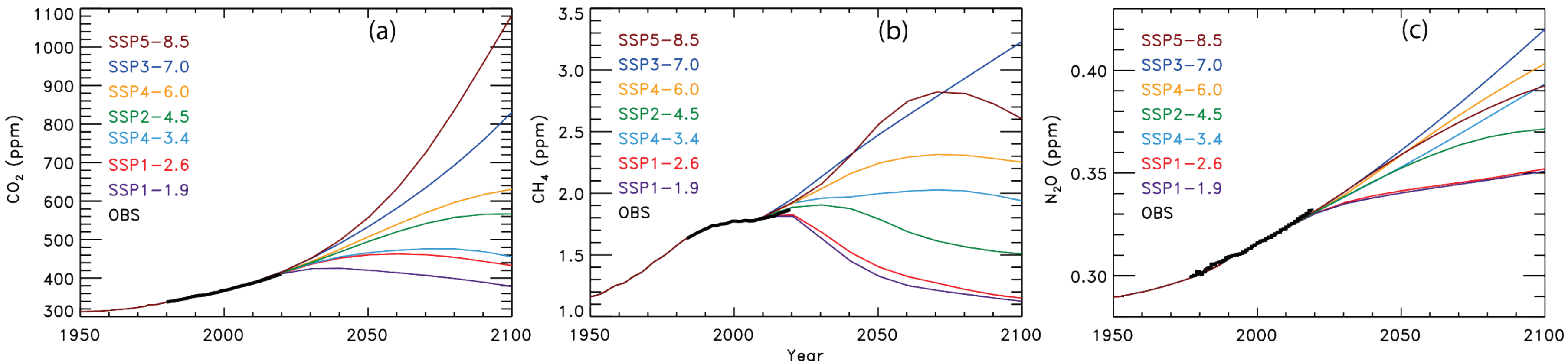
based on data from the Japanese 55 year re-analysis project

https://jra.kishou.go.jp/JRA-55/index_en.html

SOLID CIRCLES denote local maxima in PFP relative to a trend line

SSP: Shared Socioeconomic Pathway Scenarios Will Drive Upcoming IPCC Report

Climate Model Input



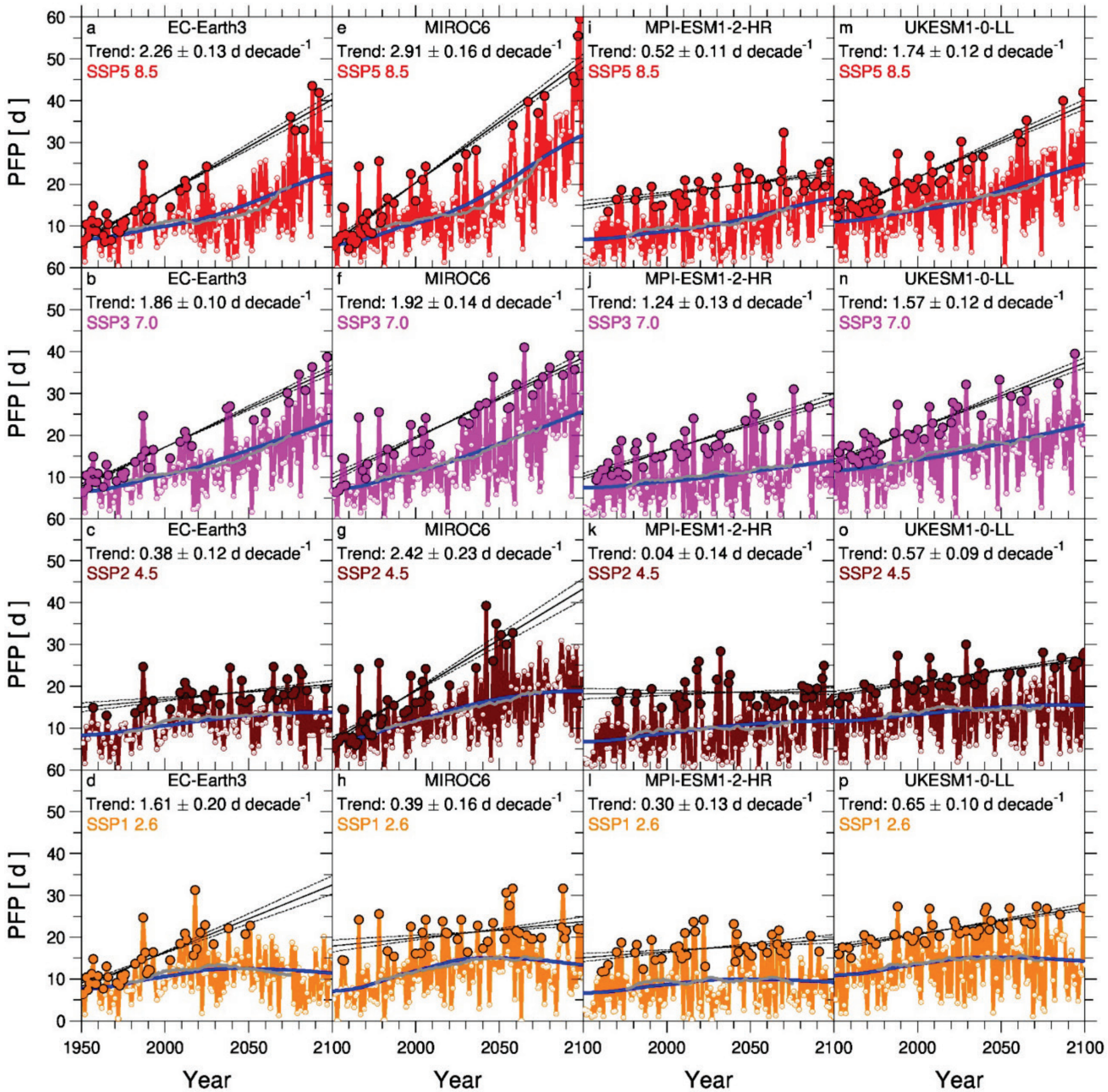
McBride et al., *Earth System Dynamics*, submitted, 2020

Number before dash represents base narrative and number after dash represents W m^{-2} RF of climate at end of century

Tendency for Colder Arctic Winters Getting Colder Drive by Rising GHGs

Climate Model Output

PFP is PSC Formation Potential



von der Gathen, *Nature Communications*, submitted, 2020

Next Week

Tuesday

- **No AT, because reading is "light" & Problem Set #3 is due**
- **Please get started on Problem Set 3 prior to Mon evening**

Thursday

- **Will hold review of Lectures 10 to 17, in preparation for second exam to be held on Tuesday, 17 November**