Review of Lectures 10 to 17 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



https://www.videoblocks.com/video/earth-sunset-spacewalk-view-from-space-station-r7dydlcsgjd23vml0

12 November 2020

1

Announcements: Class

Second Exam

Second exam which will be on-line, open book, open web, <u>that everyone will take during normal class</u> <u>hour on Tues, 17 Nov, from 2:02 to 3:17 pm.</u>

However, please note that:

a) the exam will focus on a series of question that you can only answer properly in the limited time IF you are already familiar with the contents of each lecture;

b) there will be either minimal or no calculations on the exam, the vast majority of the exam will be qualitative rather than quantitative

c) if you have been doing all of the readings, answering the ATs based on a comprehensive understanding of the readings, and retaining knowledge from the readings and exams, as solidified by consistently high scores of the learning out come quizzes, then you'll be in great shape for the first exam. On the other hand, if you have been skimming the readings, doing the bare minimum to answer the ATs, and not completing the learning outcome quizzes, you will need to impart greater effort to prepare for the exam, in order to do well.

d) by "open web", I mean you are allowed to search for information on the Web. You absolutely, positively are not allowed to conduct any on-line chats, or solicit help from an on-line assistance program of any sort.

Announcements: Outside of Class

Today, 12 Nov: AOSC Weekly Seminar (3:30 pm)

Dr. Dylan Jones, University of Toronto

Summertime ozone in North America: Isolating weather-driven ozone pollution events and evaluating trends in precursor emissions

Air pollution regulations have led to dramatic reductions in emissions of air quality pollutants in North America, and thus improvements in air quality, during the past three decades. However, ozone pollution episodes remain an issue in some regions of North America. Furthermore, there are uncertainties in the trend in emissions of nitrogen oxides (NOx), a key ozone precursor. In this talk I will examine the link between summertime ozone pollution episodes and large-scale atmospheric circulation patterns and present an analysis isolating the weather-driven component of ozone pollution episodes. I will also review the discrepancies in recent trends of emissions of NOx in the United States and discuss the use of a deep learning model to evaluate the consistency of the reported trends in NOx emissions with observations of surface ozone.

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

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

Learning Outcome Quizzes



ACC 2020 Fall, Lecture 12 Results

Filter by name / partition by tag / group by na

Average Score	Average Time	
77%	0:08:57	
60% to 100%	0:00:55 to 0:28:25	

<u>Name</u> ▲	Score	Started On	Finished On	Time
	80% (4/5)	2020-10-27 10:11 PM	2020-10-27 10:13 PM	0:02:01
	80% (4/5)	2020-10-23 1:46 PM	2020-10-23 1:51 PM	0:05:20
	80% (4/5)	2020-10-26 7:13 PM	2020-10-26 7:16 PM	0:03:50
	60% (3/5)	2020-10-23 2:52 PM	2020-10-23 3:01 PM	0:09:17
	80% (4/5)	2020-11-11 12:03 PM	2020-11-11 12:18 PM	0:13:00
	60% (3/5)	2020-10-31 10:11 AM	2020-10-31 10:39 AM	0:28:22
	100% (5/5)	2020-10-31 10:40 AM	2020-10-31 10:41 AM	0:00:53

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Importance of Radicals

- With a few exceptions, the only reactions between molecules that proceed at appreciable rates are those involving at least one radical
- Radicals require significant energy to form: a bond must be broken
- Radical formation is tied to absorption of photons that "photodissociate" a compound, leading to radical formation

Initiation

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Atmospheric Radiation

 Solar irradiance (downwelling) at top of atmosphere occurs at wavelengths between ~200 and 2000 nm (~5750 K "black body" temperature)



• Absorption and photodissociation in the UV occurs due to changes in the electronic state (orbital configuration) of molecules

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Absorption Cross Section of O₂



From Brasseur & Solomon, Aeronomy of the Middle Atmosphere, 1986

- O_2 can not dissociate longward of ~250 nm
- All of the absorption shown above is dissociative (e.g., leads to production of two O atoms)
- Structure in the O₂ cross section is related to whether the initial transition involves an unbound electronic state (smooth) or involves a specific vibrational level of an electronic state (banded, due to requirement of specific quanta of energy)

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Optical Depth of O₂ Absorption

Recall the *Beer-Lambert Law*:

 $F(z,\lambda) = F_{TOA}(\lambda) e^{-\tau(z,\lambda)}$ (TOA : Top of Atmosphere)

where:

$$\tau(z, \lambda) = m \int_{z}^{\infty} \sigma_{\lambda} [C] dz' \qquad (\tau: optical depth)$$
$$\int_{0}^{\infty} [O_{2}] dz' \approx 4 \times 10^{24} \text{ molecules/cm}^{2}$$

Also:

O_2 Optical Depth for $\theta = 0^\circ$, $z = 0$ km			
	$\sigma_{\rm max}({\rm cm}^2)$	τ (0 km)	$e^{-\tau (0 \text{ km})}$
Schumann-Runge Continuum	10^{-17}	4×10^7	0.
Schumann-Runge Bands	10^{-20}	4×10^4	0.
	3×10^{-23}	120	7.6×10^{-53}
Herzberg Continuum	10 ⁻²³	40	4.2×10^{-18}

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Optical Depth of O₃ Absorption

A typical mid-latitude column abundance for O_3 is 300 Dobson units (DU):

1 DU = 2.687×10^{16} molecules/cm²; 300 DU = 8×10^{18} molecules/cm²

Aside:

$$\frac{\text{Column O}_3}{\text{Column Air}} = \frac{8 \times 10^{18}}{2 \times 10^{25}} = 0.4 \text{ parts per million} \implies \text{Ozone is a trace species !}$$

O_3 Optical Depth for $\theta = 0^\circ$, $z = 0$ km				
	$\sigma_{\rm max}({\rm cm}^2)$	τ (0 km)	$e^{-\tau (0 \text{ km})}$	O_3 Column, $\tau = 1.0$
Hartley (~220 to 280 nm)	10^{-17}	80	1.8×10^{-35}	3.7 DU
Huggins (~310 to 330 nm)	10 ⁻¹⁹	0.8	0.45	372 DU
Chappuis (~500 to 700 nm)	3 × 10 ⁻²¹	0.024	~1.0	12,400 DU

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Solar Spectral Actinic Flux



130 ATMOSPHERIC PHOTOCHEMISTRY AND CHEMICAL KINETICS

From DeMore et al., *Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling*, Evaluation No. 11, 1994.

From Seinfeld and Pandis, Atmospheric Chemistry and Physics, 1998.

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Photolysis Frequency

For a specific spectral interval, the photolysis frequency (*partial J value*) of a gas is given by the product of its absorption cross section and the solar irradiance:

$$J_{gas}(z,\lambda) = Quantum_Yield(\lambda) \sigma_{gas}(\lambda,T) F(z,\lambda)$$

Units: s⁻¹ nm⁻¹

The total *photolysis frequency* (*J value*) is found by integrating $J_{gas}(z,\lambda)$ over all wavelengths for which the gas photodissociates:

$$J_{gas}(z) = \int_{\lambda_{min}}^{\lambda_{max}} J_{gas}(z, \lambda) \, d\lambda \qquad \text{Units: } s^{-1}$$

Rate of Reaction =
$$\frac{dO_3}{dt} = J$$
 [O₃]; Units of J are s⁻¹

More precisely, calculations of photolysis frequencies consider the "spectral actinic flux", which represents the amount of available photons integrated over all angles, rather than "solar irradiance". These two quantities differ because of scattering of solar radiation by gases and aerosols, and reflection of radiation by clouds and the surface.

NO₂ Photolysis



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$O_3 \rightarrow O(^1D)$ Photolysis



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Bimolecular Gas Phase Reactions



Bimolecular Gas Phase Reactions



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Bimolecular Gas Phase Reactions



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Tropospheric Ozone Production versus NO Production of Tropospheric O₃ limited by: _____ ? As NO_x rises: [HO₂] falls faster than [NO] rises,

leading to a decrease in the value _



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Tropospheric Ozone Production versus NO Production of Tropospheric O₃ limited by: $k[HO_2][NO] + \Sigma k[RO_2][NO]$ As NO_x rises:

[HO₂] falls faster than [NO] rises, leading to a decrease in the value _



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Tropospheric Ozone Production versus NO Production of Tropospheric O₃ limited by: $k[HO_2][NO] + \Sigma k[RO_2][NO]$ As NO_x rises:

[HO₂] falls faster than [NO] rises,

leading to a decrease in the value the production rate of O₃



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Tropospheric Ozone Production versus NO_x and VOCs

Ridge: local maximum for O₃ that separates the NOx-limited regime from and VOC limited regime



Figure: http://www-personal.umich.edu/~sillman/ozone.htm

An important discovery in the past decade is that the focus on hydrocarbon emission controls to combat O_3 pollution may have been partly misdirected. Measurements and model calculations now show that O_3 production over most of the United States is primarily NO_x limited, not hydrocarbon limited. The early models were in error in part because they underestimated emissions of hydrocarbons from automobiles, and in part because they did not account for natural emission of biogenic hydrocarbons from trees and crops.

Jacob, Chapter 12, Introduction to Atmospheric Chemistry, 1999

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Day-to-day meteorology (weather!) affects severity and duration of pollution episodes



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Day-to-day meteorology (weather!) affects severity and duration of pollution episodes



http://www.mde.state.md.us/assets/document/BJH%20-%20Basics%20on%20Ozone%20Transport.ppt

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Significant Improvements in Local Air Quality since early 1980s



http://www.mde.state.md.us/programs/Air/AirQualityMonitoring/Pages/SeasonalReports.aspx

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Nitrate Deposition (see Fig 6.12)

1986

2016



National Atmospheric Deposition Program/National Trends Network http://nadp.isws.illinois.edu



National Atmospheric Deposition Program/National Trends Network http://nadp.slh.wisc.edu

http://nadp.slh.wisc.edu/data/animaps.aspx

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Sulfate Deposition (see Fig 6.12)

1986

2016



National Atmospheric Deposition Program/National Trends Network http://nadp.isws.illinois.edu



National Atmospheric Deposition Program/National Trends Network http://nadp.slh.wisc.edu

http://nadp.slh.wisc.edu/data/animaps.aspx

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pH of rain samples (see Fig 6.11)

2016

1986



National Atmospheric Deposition Program/National Trends Network http://nadp.isws.illinois.edu



National Atmospheric Deposition Program/National Trends Network http://nadp.slh.wisc.edu

http://nadp.slh.wisc.edu/data/animaps.aspx

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US Trends: NO₂ and SO₂



Krotkov et al., ACP, 2016

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China Trends: NO₂ and SO₂



Krotkov et al., ACP, 2016

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Stratospheric Ozone: Chapman Chemistry

- Production of O₃ initiated when O₂ is photodissociated by UV sunlight
- O_3 formed when resulting O atom reacts with O_2 :

$$h\nu + O_2 \rightarrow O + O \qquad (1)$$

$$O + O_2 + M \rightarrow O_3 + M \qquad (2)$$

• O₃ removed by photodissociation (UV sunlight) or by reaction with O :

This reaction sequence was first worked out in the 1930s by Sidney Chapman, an English mathematician and geophysicist

Chapman Chemistry

- The cycling between O and O₂ (rxns 2 and 3) occurs *much* more rapidly than leakage into (rxn 1) or out of the system (rxn 4)
- The sum O + O₃ is commonly called *"odd oxygen"*



Rxn (1) produces two *odd oxygen* molecules Rxn (4) consumes two *odd oxygen* molecules

and reactions 2 and 3 recycle *odd oxygen* molecules



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.

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

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One Atmosphere – One Photochemistry Stratosphere Troposphere

HO₂ formation: $OH + O_3 \rightarrow HO_2 + O_2$ HO₂ loss: $HO_2 + O_3 \rightarrow OH + 2 O_2$ Net: $O_3 + O_3 \rightarrow 3 O_2$ HO₂ formation: OH + CO $\xrightarrow{O_2}$ HO₂ + CO₂ HO₂ loss: HO₂ + NO \rightarrow OH + NO₂ Followed by: NO₂ + hv \rightarrow NO +O O+ O₂ + M \rightarrow O₃ + M Net: CO + 2 O₂ \rightarrow CO₂ + O₃



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Rate HO₂ Formation = $k_{OH+O3} \times [OH][O_3] + k_{OH+CO} \times [OH][CO]$ Rate HO₂ Loss = $k_{HO2+O3} \times [HO_2][O_3] + k_{HO2+NO} \times [HO_2][NO]$ HO₂ formation: OH + CO \rightarrow HO₂ + CO₂ HO₂ loss: HO₂ + NO \rightarrow OH + NO₂ Followed by: NO₂ + hv \rightarrow NO +O O+ O₂ + M \rightarrow O₃ + M Net: CO + 2 O₂ \rightarrow CO₂ + O₃

 $\begin{aligned} & \textbf{Rate HO}_{2} \ \textbf{Formation} = \textbf{k}_{OH+O3} \times [OH][O_{3}] + \textbf{k}_{OH+CO} \times [OH][CO] \\ & \textbf{Rate HO}_{2} \ \textbf{Loss} = \textbf{k}_{HO2+O3} \times [HO_{2}][O_{3}] + \textbf{k}_{HO2+NO} \times [HO_{2}][NO] \end{aligned}$



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Montreal Protocol Has Banned Industrial Production of CFCs & Other ODS

Projections Based on 2018 World Meteorological Organization

Scientific Assessment of Ozone Depletion Report



https://www.esrl.noaa.gov/csd/assessments/ozone/2018

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Chlorine Abundance, Mid-Latitude Stratosphere



Fig Q7-2, WMO/UNEP Twenty QAs Ozone

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Chlorine Source Gases



Fig Q6-1, WMO/UNEP Twenty QAs Ozone

Time series of chlorine content of organic halocarbons that reach the stratosphere. Past values based on direct atmospheric observation. Future values based on projections that include the lifetime for removal of each halocarbon.

Table 6-4, WMO/UNEP 2018

Gas	Atmospheric Lifetime (years)	Ozone Depletion Potential (ODP) ^b		
Halogen Source Gases				
Chlorine Gases				
CFC-11 (CCl ₃ F)	52	1		
Carbon tetrachloride (CCl ₄)	32	0.87		
CFC-113 (CCl ₂ FCClF ₂)	93	0.81		
CFC-12 (CCl ₂ F ₂)	102	0.73		
Methyl chloroform (CH ₃ CCl ₃)	5.0	0.14		
HCFC-141b (CH ₃ CCl ₂ F)	9.4	0.102		
HCFC-142b (CH ₃ CCIF ₂)	18	0.057		
HCFC-22 (CHF ₂ Cl)	12	0.034		
Methyl chloride (CH ₃ Cl)	0.9	0.015		
Bromine Gases				
Halon-1301 (CBrF ₃)	65	15.2		
Halon-1211 (CBrCIF ₂)	16	6.9		
Methyl bromide (CH ₃ Br)	0.8	0.57		
Hydrofluorocarbons (HFCs	\$)			
HFC-23 (CHF ₃)	228	0		
HFC-143a (CH ₃ CF ₃)	51	0		
HFC-125 (CHF ₂ CF ₃)	30	0		
HFC-134a (CH ₂ FCF ₃)	14	0		
HFC-32 (CH ₂ F ₂)	5.4	0		
HFC-152a (CH ₃ CHF ₂)	1.6	0		
HFO-1234yf (CF ₃ CF=CH ₂)	0.03	0		

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Mid-Latitude Ozone Depletion

Total column ozone anomaly is deseasonalized, cosine latitude weighted average of total column ozone collected between 60°S and 60°N, relative to the mean total column abundance over the entire time period.





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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.

POLAR OZONE LOSS

- <u>COLD TEMPERATURES</u> → POLAR STRATOSPHERIC CLOUDS (<u>PSCs</u>)
- REACTIONS ON PSC SURFACES LEAD TO ELEVATED <u>CIO</u>

HCI + CINO₃ → Cl₂ (gas) + HNO₃ (solid) CINO₃ + H₂O → HOCI + HNO₃ Cl₂ + SUNLIGHT + O₃ → CIO HOCI + SUNLIGHT + O₃ → CIO HNO₃ SEDIMENTS (PSCs fall due to gravity)

- ELEVATED CIO + <u>SUNLIGHT</u> DESTROYS O_3
- BrO : REACTION PARTNER FOR CIO \Rightarrow Additional O₃ Loss



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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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Minimum Temperature: NH and SH



Fig Q9-1, WMO/UNEP Twenty QAs Ozone

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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: <u>https://www.youtube.com/watch?v=4aq_F9Ma0DQ</u>

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Antarctic Vortex Minimum Temperature: 2018-2019



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

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Antarctic Vortex Minimum Temperature: 2019-2020



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

Copyright © 2020 University of Maryland.

Antarctic Vortex Minimum Temperature: 2019-2020



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

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Arctic Ozone Loss Varies as a function of <u>PSC Formation Potential</u>



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

$$PFP = \int_{16 \text{ Nov}}^{17 \text{ Apr}} \frac{V_{PSC}(t)}{V_{VORTEX}(t)} dt \qquad PFP \text{ is } \underline{PSC} \ \underline{F} \text{ ormation } \underline{P} \text{ otential}$$

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

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Arctic Ozone 2020 in Context of Prior Years



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

Arctic Temperature: Mar 2019



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

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Arctic Temperature: Mar 2020



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

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Arctic Ozone: 2019 and 2020



0 100 200 300 400 500 600 700 Total Ozone (Dobson units)

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

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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

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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



von der Gathen, Nature Communications, submitted, 2020

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Future Ozone: ODSs, CO₂, CH₄ and N₂O



Fig Q20-3, 20 QAs, WMO (2019)

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Longstreth et al., J. of Photochemistry and Photobiology B, 46, 20–39, 1998.

See also Slaper *et al.*, Estimates of ozone depletion and skin cancer incidence to examine the Vienna Convention achievements, *Nature*, *384*, 256–258, 1996, who state:

The no-restrictions and Montreal Protocol scenarios produce a runaway increase in skin cancer incidence, up to a quadrupling and doubling, respectively, by year 2100.

Announcements: Class

Second Exam

Second exam which will be on-line, open book, open web, *that everyone will take during normal class hour on Tues, 17 Nov, from 2:02 to 3:17 pm.*

However, please note that:

a) the exam will focus on a series of question that you can only answer properly in the limited time IF you are already familiar with the contents of each lecture;

b) there will be either minimal or no calculations on the exam, the vast majority of the exam will be qualitative rather than quantitative

c) if you have been doing all of the readings, answering the ATs based on a comprehensive understanding of the readings, and retaining knowledge from the readings and exams, as solidified by consistently high scores of the learning out come quizzes, then you'll be in great shape for the first exam. On the other hand, if you have been skimming the readings, doing the bare minimum to answer the ATs, and not completing the learning outcome quizzes, you will need to impart greater effort to prepare for the exam, in order to do well.

d) by "open web", I mean you are allowed to search for information on the Web. You absolutely, positively are not allowed to conduct any on-line chats, or solicit help from an on-line assistance program of any sort.