



Interactive chemistry for E3SM: capabilities and initial results

Qi Tang (LLNL)

Michael Prather (UCI), Juno Hsu (UCI)

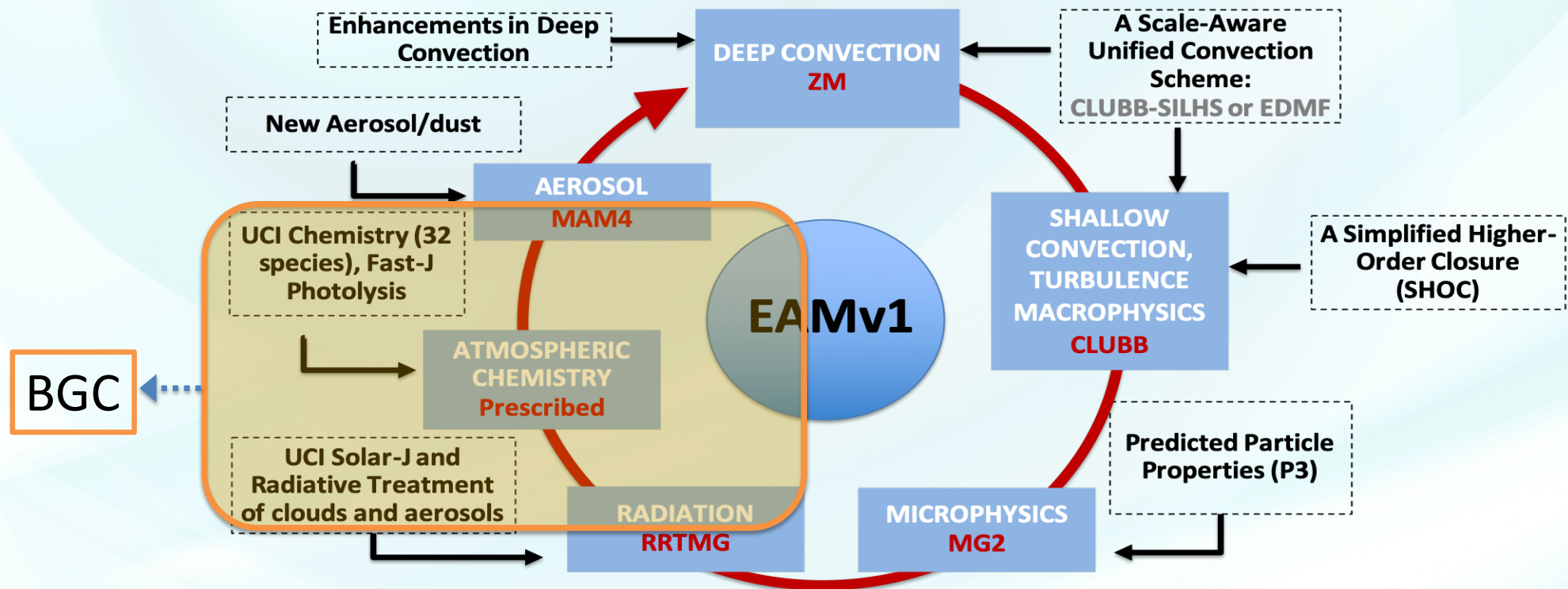
Philip Cameron-Smith (LLNL)

E3SM BGC webinar, May 25, 2021

Acknowledgments: E3SM team; Compy & NERSC computing resources

NGD-Atmospheric Physics for E3SMv3

Goals: 1) Reduce model errors in v1/v2; 2) Enhance model capability for future climate projections



Atmospheric chemistry and radiation

Ability to project future GHGs (CH_4 , N_2O , O_3) based on emission scenarios and climate change by implementing the UC Irvine (UCI) chemistry mechanism

- Support BGC and aerosol chemistry including short-lived climate forcers

Starting point

E3SMv1 chemistry (O3v1)

- Strat: linearized interactive ozone (Linoz v2); Trop: prescribed oxidants for aerosols
- Misses tropopause (TPP) variability, esp. at the jet.

E3SMv2 chemistry (O3v2) (Tang et al., 2021)

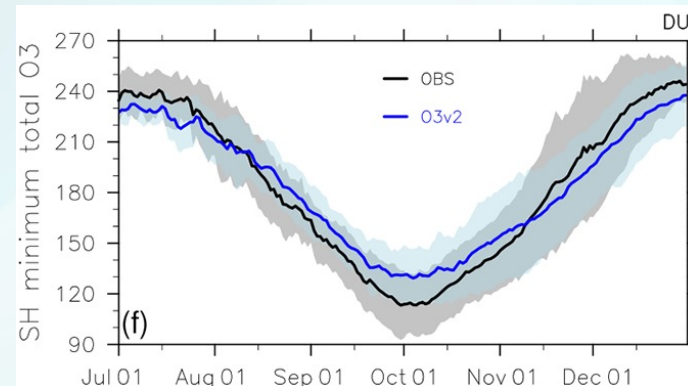
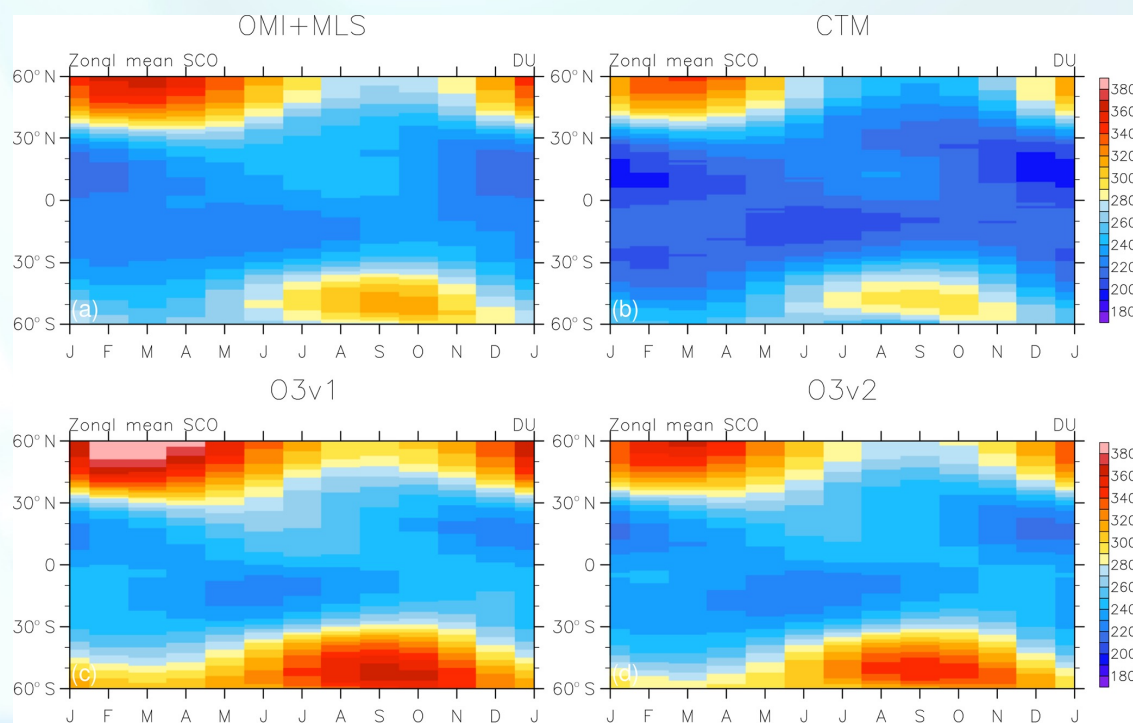
- Trop O_3 becomes a passive tracer with lower boundary sink.
- Maintains the naturally sharp O_3 gradient across TPP
- Able to diagnose stratosphere troposphere exchange (STE) of ozone

Recent E3SM publications

Evaluation of the interactive stratospheric ozone (O3v2) module in the E3SM version 1 Earth system model

Qi Tang¹, Michael J. Prather², Juno Hsu², Daniel J. Ruiz², Philip J. Cameron-Smith¹, Shaocheng Xie¹, and Jean-Christophe Golaz¹

Geosci. Model Dev., 14, 1219–1236, 2021
<https://doi.org/10.5194/gmd-14-1219-2021>



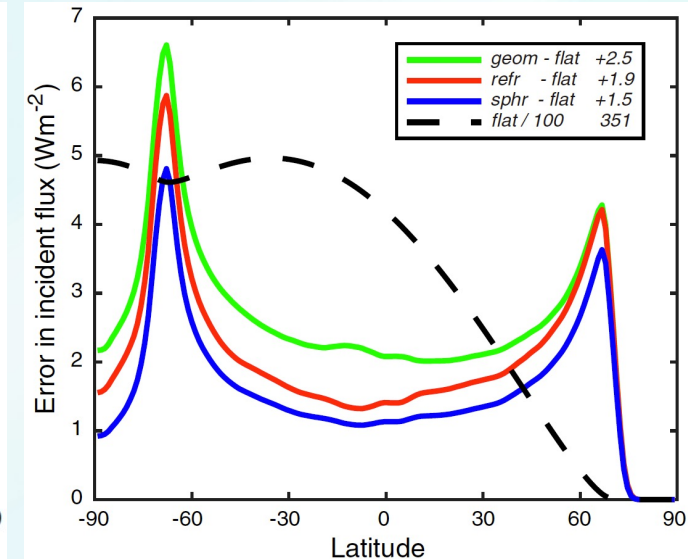
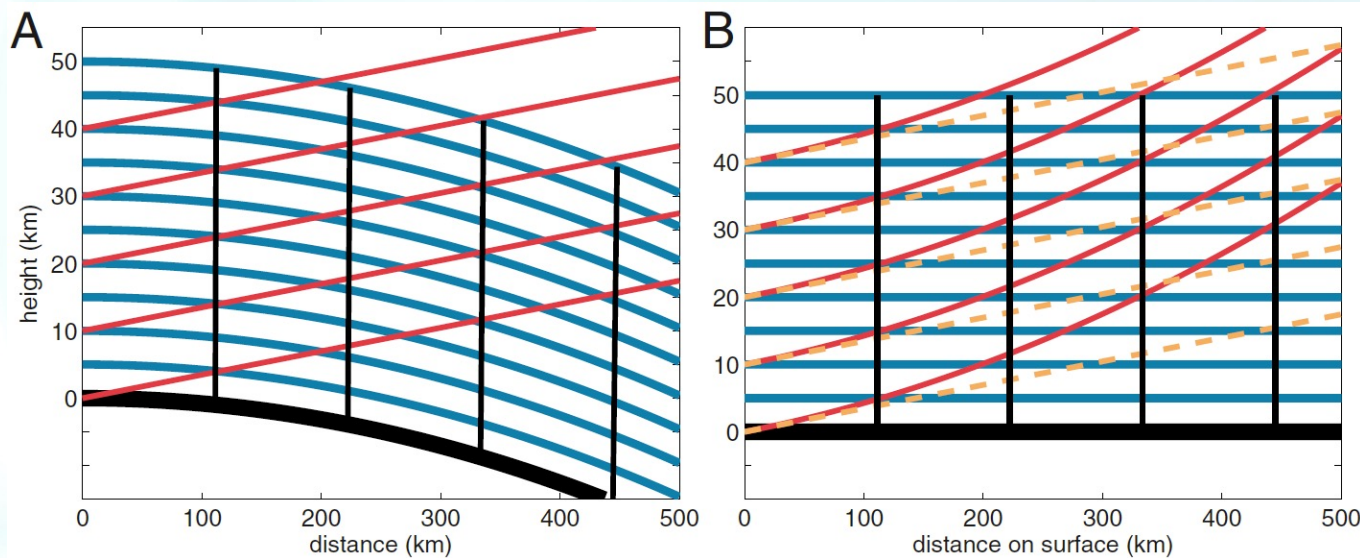
Stratospheric column ozone (SCO) is reasonably captured by Linoz v2, including the Antarctic ozone hole.

A round Earth for climate models

Michael J. Prather^{a,1,2} and Juno C. Hsu^{a,1}

19330–19335 | PNAS | September 24, 2019 | vol. 116 | no. 39

www.pnas.org/cgi/doi/10.1073/pnas.1908198116



- Flat atmosphere approximation leads to regional errors in solar heating at rates comparable to the climate forcing by greenhouse gases and aerosols.
- Included in the Fast-J code in E3SM (not enabled for radiation transfer)

Assessing Uncertainties and Approximations in Solar Heating of the Climate System

Juno C. Hsu¹  and Michael J. Prather¹ 

JAMES | Journal of Advances in
Modeling Earth Systems
10.1029/2020MS002131

Assessment of errors and uncertainties in solar heating codes with recommendations

Table 1

Errors in the Three Primary Components of the Solar Radiation Budget (Reflected Sunlight, Atmospheric Absorption, Surface Absorption in $W m^{-2}$) for a Range of Approximations in the Radiative Transfer Models

row	Models	Error being estimated	Mean difference (Wm ⁻²)			RMS difference (Wm ⁻²)		
			refl.	Atm.	Surf.	refl.	Atm.	Surf.
Spectroscopy errors for infrared and visible gas absorption with clear sky								
R1	B1–B0	SJ/CLIRAD v. SJ	+0.94	−5.68	+4.73	1.4	8.4	7.0
R2	B2–B0	SJ/LLNL v. SJ	+0.53	−7.66	+7.13	1.2	12.0	11.0
R3	B3–B0	SJ/RRX v. SJ	+0.03	−0.24	+0.21	0.0	0.4	0.3
R4	RR0–B0	AER4.0 v. SJ	+1.30	−0.47	-0.82	1.9	0.8	1.4
R5	B4–B0	SJ/hrv v. SJ	+0.34	−0.46	+0.12	-	-	-
R6	B5–B0	SJ/2S versus SJ	+0.31	−0.53	+0.22	0.4	0.4	0.2
Wavelength resolution of cloud absorption (<i>MAX-COR overlap and QCAs, no IR gas absorption</i>)								
R7	C1–C0	SJ/CLIRAD v. SJ/66b	−2.14	+3.85	−1.70	8.7	10.4	3.1
R8	C2–C0	SJ/RRTM v. SJ/66b	−1.14	+1.68	−0.51	2.2	3.2	1.1



Current status (ongoing work)

Implemented on E3SM GitHub branches

- Interactive chemistry ready to link with BGC and aerosols
 - Strat: Linoz v3; Trop: chemUCI
 - BGC methane experiment and other scientific applications
- Fast-J photolysis rates
 - Replace the look-up table and is coupled with the super-fast chemistry

E3SMv3 tracers and reactions

Fast interactive chemistry: ~12 SYPD on 30 compy nodes (~8% additional cost)

Strat: Linoz v3

O3LNZ
CH4LNZ
N2OLNZ
NOYLNZ

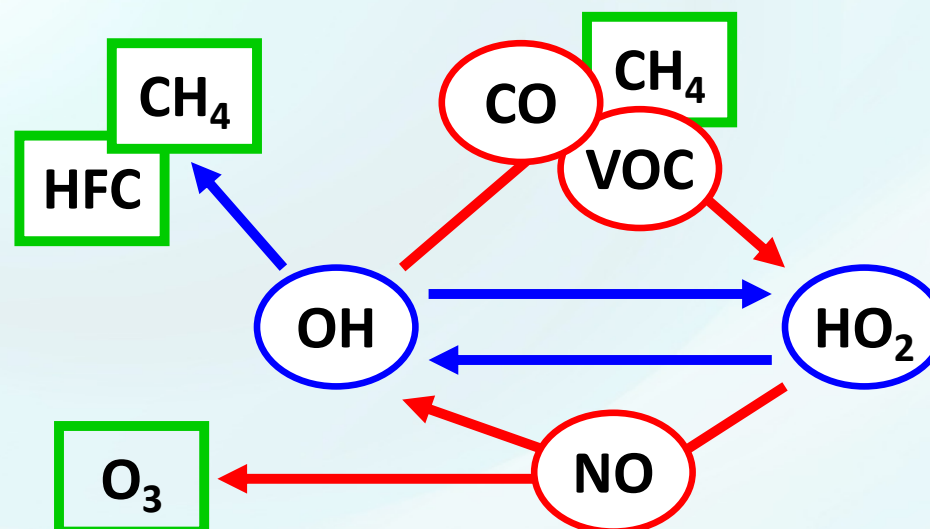
- Identify TPP: E90
- Reactions: https://github.com/E3SM-Project/E3SM/blob/tangq/atm/UCI-chem/components/eam/chem_proc/inputs/pp_chemUCI_linozv3_mam4_resus_mom_so_ag_tag.in

Trop: chemUCI

O3	NO3	C2H5O2
OH	N2O5	C2H5OOH
CH4*	HNO3	CH3CHO
HO2	PAN	CH3CO3
H2O2	CO	ISOP (C5H8)
CH2O	C2H6	ISOPO2
CH3OO	C3H8	CH3COCH3
CH3OOH	C2H4	MVKO2 (C4H7O4)
NO	ROHO2 (C2H5O3)	MVKMACR (Methyvinylketone & Methacrolein)
NO2		

E3SM and chemUCI

How do **tropospheric pollutants** interact with **greenhouse gases**?



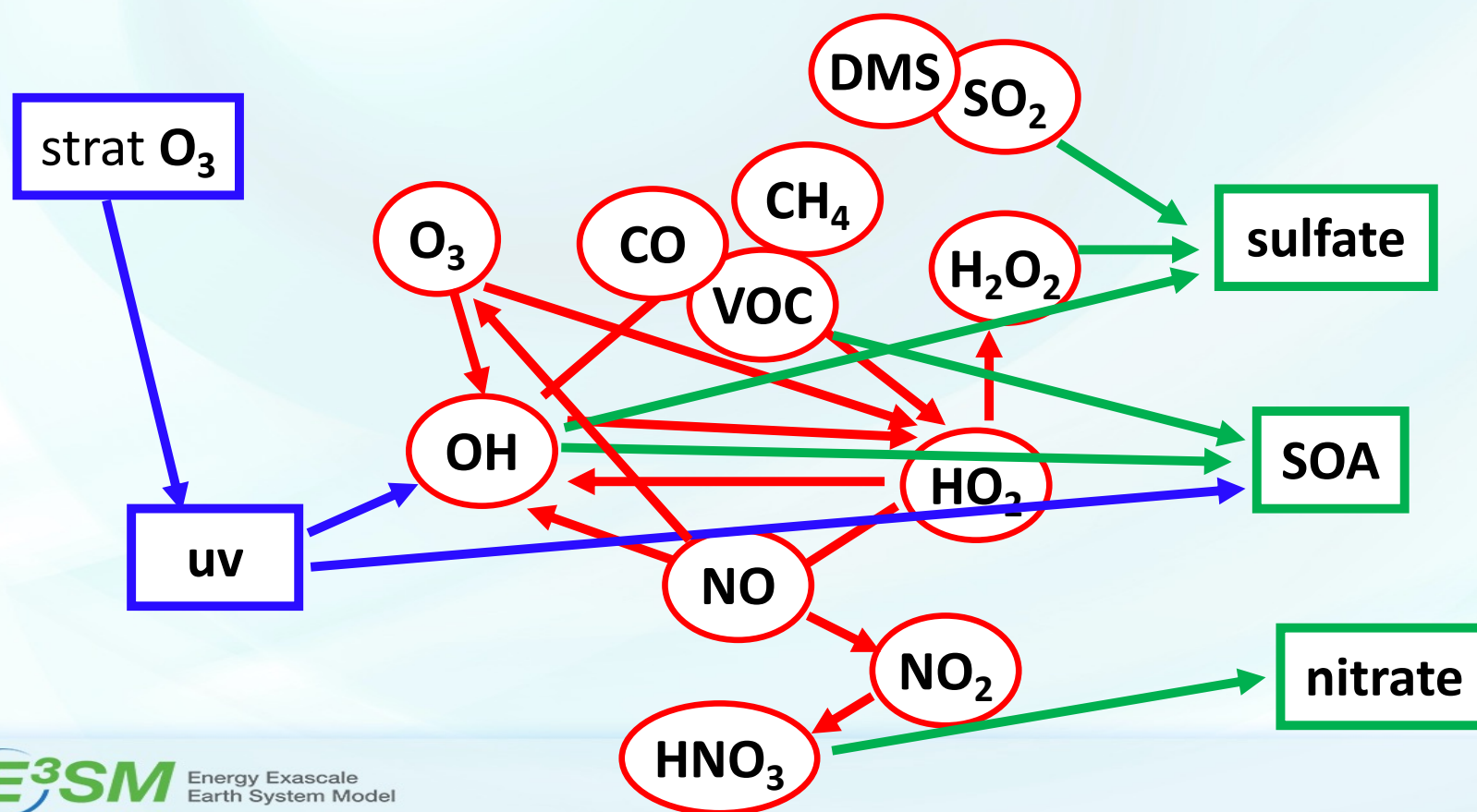
CO , VOC , NO_x ($=\text{NO}+\text{NO}_2$), & CH_4 control

Tropospheric Chemistry (OH , HO_2)

which is the sink for CH_4 & HFCs ; the source for O_3

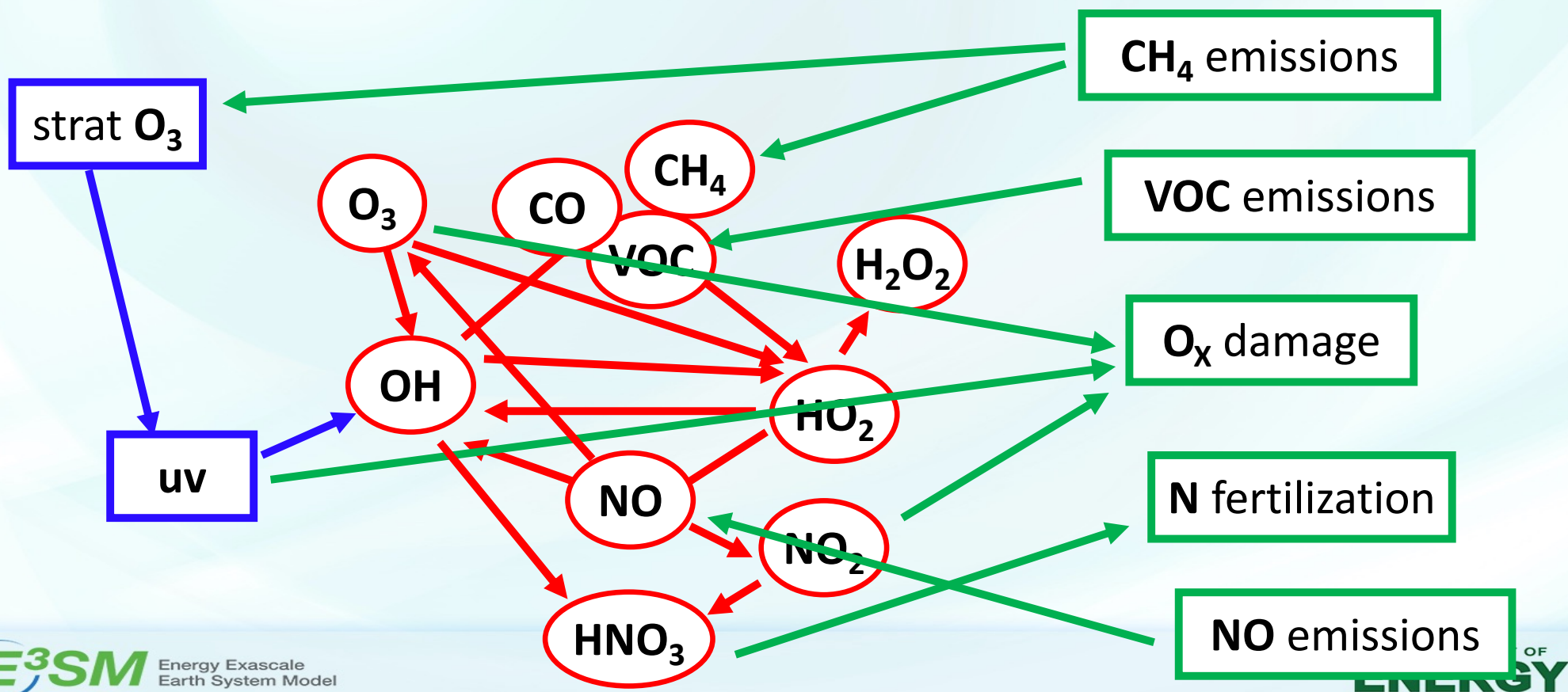
E3SM and chemUCI and Linoz v3

How does *stratospheric* & *tropospheric chemistry* interact with *aerosols*?



E3SM and chemUCI and Linoz v3

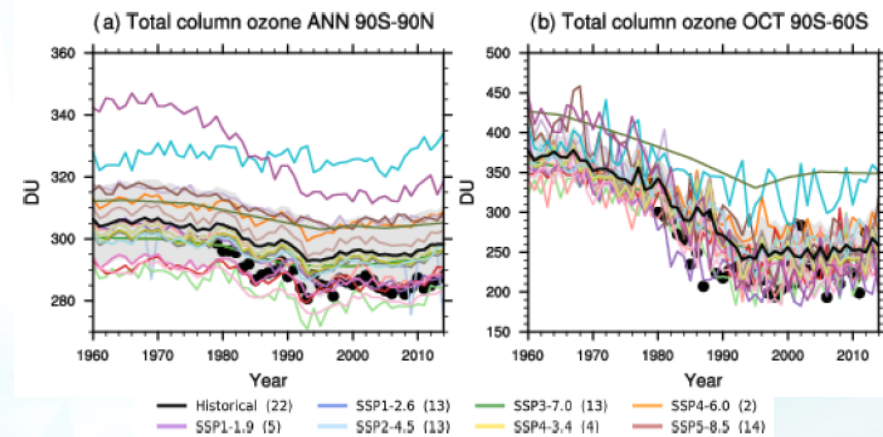
How does *stratospheric/tropospheric chemistry* interact with *BGC*?



What is being asked of ESMs for CMIP6?

- What drives chemUCI + Linoz v3 design
- E3SM capability

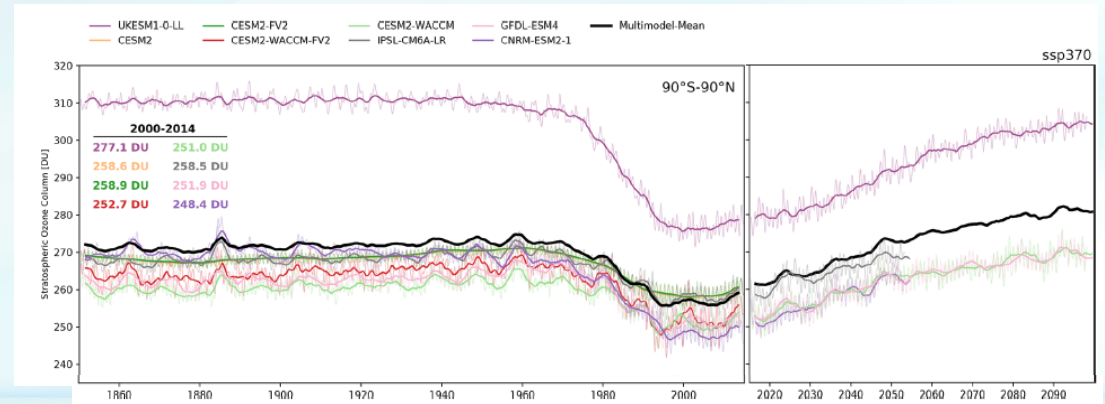
Compare Ozone Column with obs. depletion



Evaluating stratospheric ozone and water vapour changes in CMIP6 models from 1850 to 2100

James Keeble^{1,2}, Birgit Hassler³, Antara Banerjee^{4,5}, Ramiro Checa-Garcia⁶, Gabriel Chiodo^{7,8}, Sean Davis⁴, Veronika Eyring^{3,9}, Paul T. Griffiths^{1,2}, Olaf Morgenstern¹⁰, Peer Nowack^{11,12}, Guang Zeng¹⁰, Jiankai Zhang¹³, Greg Bodeker^{14,15}, Susannah Burrows¹⁶, Philip Cameron-Smith¹⁷, David Cugnet¹⁸, Christopher Danek¹⁹, Makoto Deushi²⁰, Larry W. Horowitz²¹, Anne Kubin²², Lijuan Li²³, Gerrit Lohmann¹⁹, Martine Michou²⁴, Michael J. Mills²⁵, Pierre Nabat²⁴, Dirk Olivie²⁶, Sungsu Park²⁷, Øyvind Seland²⁶, Jens Stoll²², Karl-Hermann Wiener²⁸, and Tongwen Wu²⁹

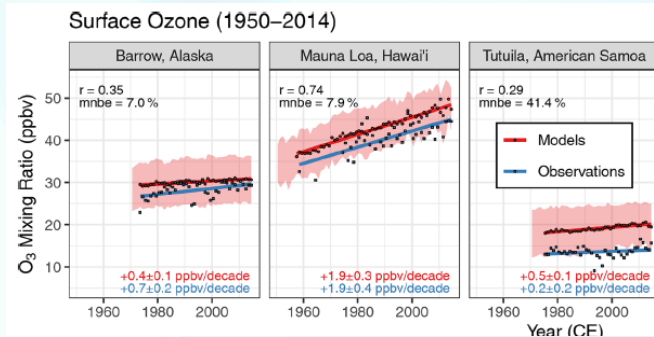
Stratospheric Ozone Column: history + projections as indicator of stratospheric climate change



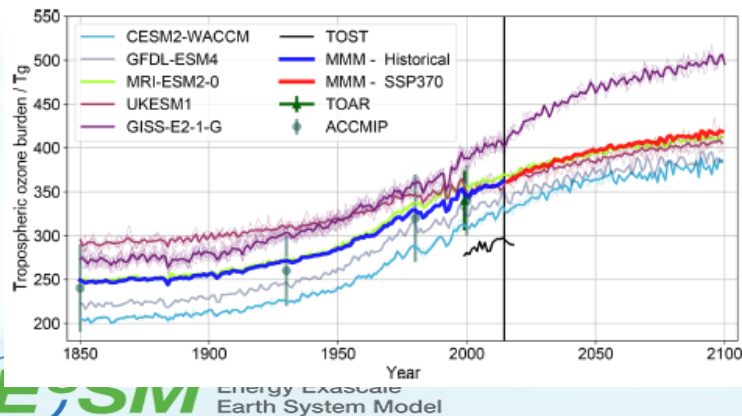
What is being asked of ESMs for CMIP6?

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- E3SM capability

Surface Ozone Trends



Tropospheric Ozone (burden) history + projection

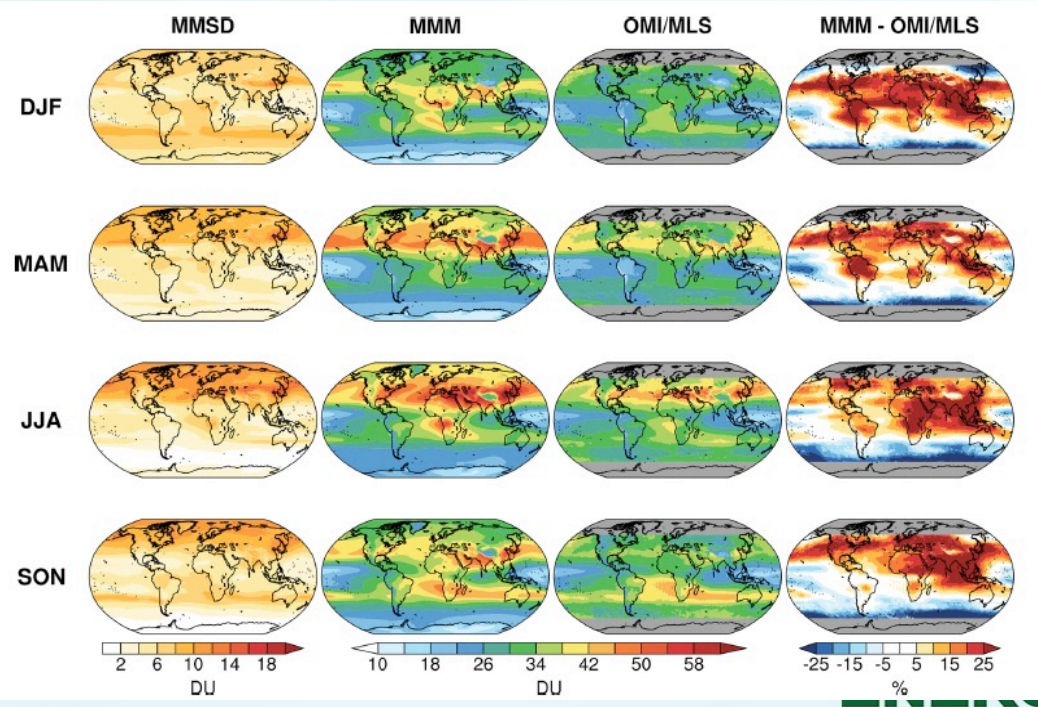


Tropospheric ozone in CMIP6 simulations

Paul T. Griffiths^{1,2,★}, Lee T. Murray^{3,★}, Guang Zeng⁴, Youngsub Matthew Shin¹, N. Luke Abraham^{1,2}, Alexander T. Archibald^{1,2}, Makoto Deushi⁵, Louisa K. Emmons⁶, Ian E. Galbally^{7,8}, Birgit Hassler⁹, Larry W. Horowitz¹⁰, James Keeble^{1,2}, Jane Liu¹¹, Omid Moeini¹², Vaishali Naik¹⁰, Fiona M. O'Connor¹³, Naga Oshima⁵, David Tarasick¹², Simone Tilmes⁶, Steven T. Turnock¹³, Oliver Wild¹⁴, Paul J. Young^{14,15}, and Prodromos Zanis¹⁶

Tropospheric Column Ozone (2005–2014)

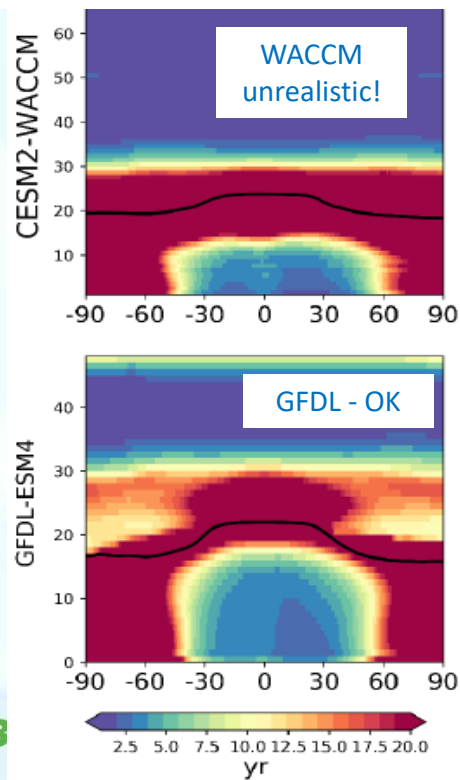
MMM = multi-model mean



What is being asked of ESMs for CMIP6?

- What drives chemUCI + Linoz v3 design
- E3SM capability

Present Day CH₄ lifetime (inverse of local loss frequency)

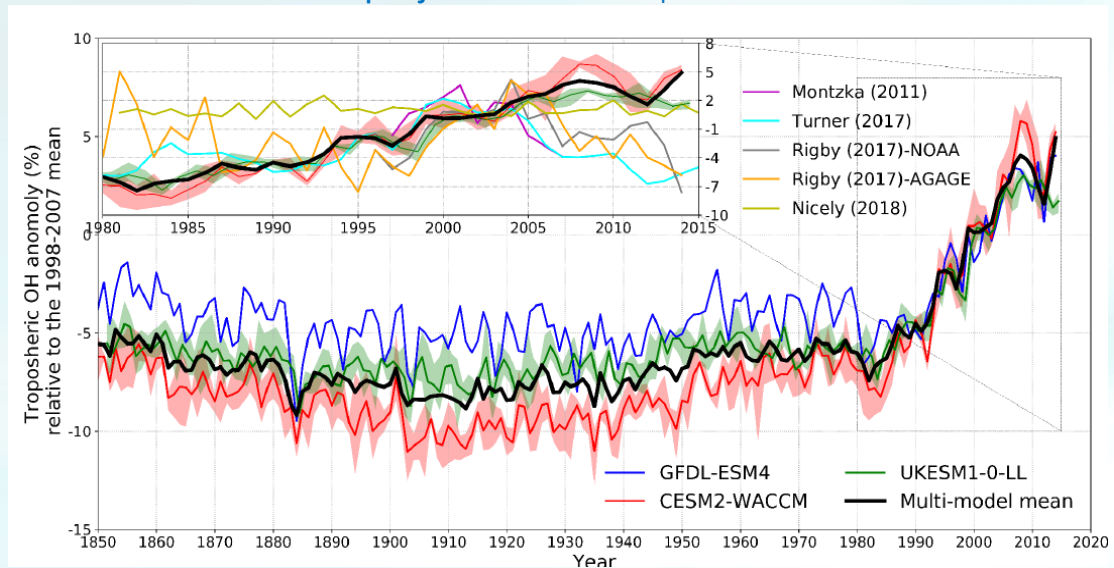


Trends in global tropospheric hydroxyl radical and methane lifetime since 1850 from AerChemMIP

David S. Stevenson¹, Alcide Zhao¹, Vaishali Naik², Fiona M. O'Connor³, Simone Tilmes⁴, Guang Zeng⁵, Lee T. Murray⁶, William J. Collins⁷, Paul Griffiths^{8,9}, Sungbo Shim¹⁰, Larry W. Horowitz², Lori Sentman², Louisa Emmons⁴

Tropospheric mean OH anomalies (related to CH₄ lifetime)

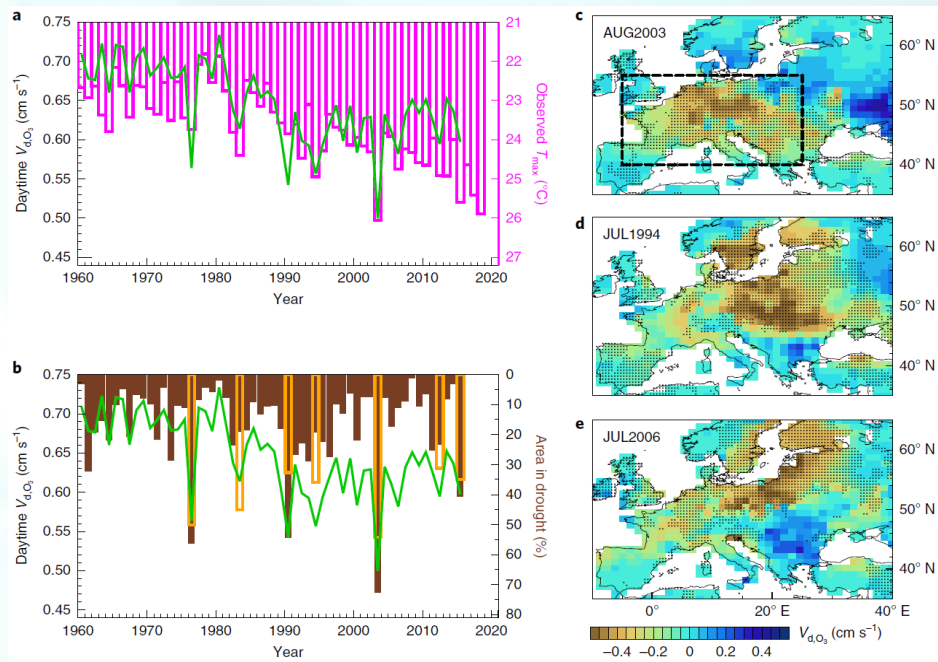
- observations for 1980-2020
- will need projections for CH₄ and HFCs



What is being asked of ESMs for CMIP6?

- What drives chemUCI + Linoz v3 design
- E3SM capability

Declining O_3 removal (V_{d,O_3}) by water-stressed vegetation



$V_{\text{deposition of } O_3}$ drops from 0.7 cm/s to 0.5 cm/s during droughts and heatwaves

Vegetation feedbacks during drought exacerbate ozone air pollution extremes in Europe

Meiyun Lin^{1,2}✉, Larry W. Horowitz², Yuanyu Xie^{1,2}, Fabien Paulot², Sergey Malyshev², Elena Shevliakova², Angelo Finco³, Giacomo Gerosa³, Dagmar Kubistin⁴ and Kim Pilegaard⁵

Reducing surface ozone to meet the European Union's target for human health has proven challenging despite stringent controls on ozone precursor emissions over recent decades. The most extreme ozone pollution episodes are linked to heatwaves and droughts, which are increasing in frequency and intensity over Europe, with severe impacts on natural and human systems. Here, we use observations and Earth system model simulations for the period 1960–2018 to show that ecosystem–atmosphere interactions, especially reduced ozone removal by water-stressed vegetation, exacerbate ozone air pollution over Europe. These vegetation feedbacks worsen peak ozone episodes during European mega-droughts, such as the 2003 event, offsetting much of the air quality improvements gained from regional emissions controls. As the frequency of hot and dry summers is expected to increase over the coming decades, this climate penalty could be severe and therefore needs to be considered when designing clean air policy in the European Union.

Leads to worse ozone pollution over Europe:
climate penalty on ozone extremes (ppb/K)
with interactive vegetation

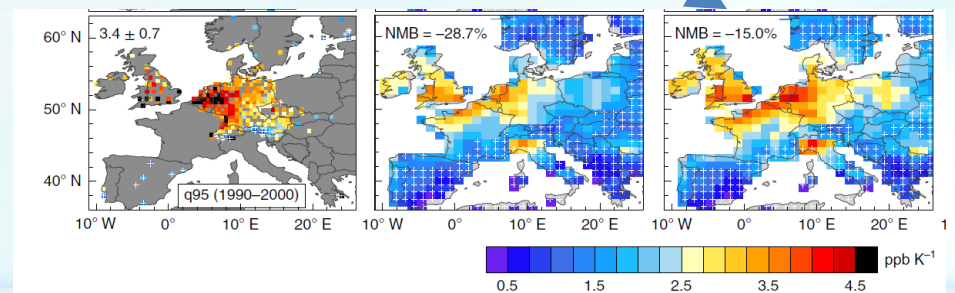
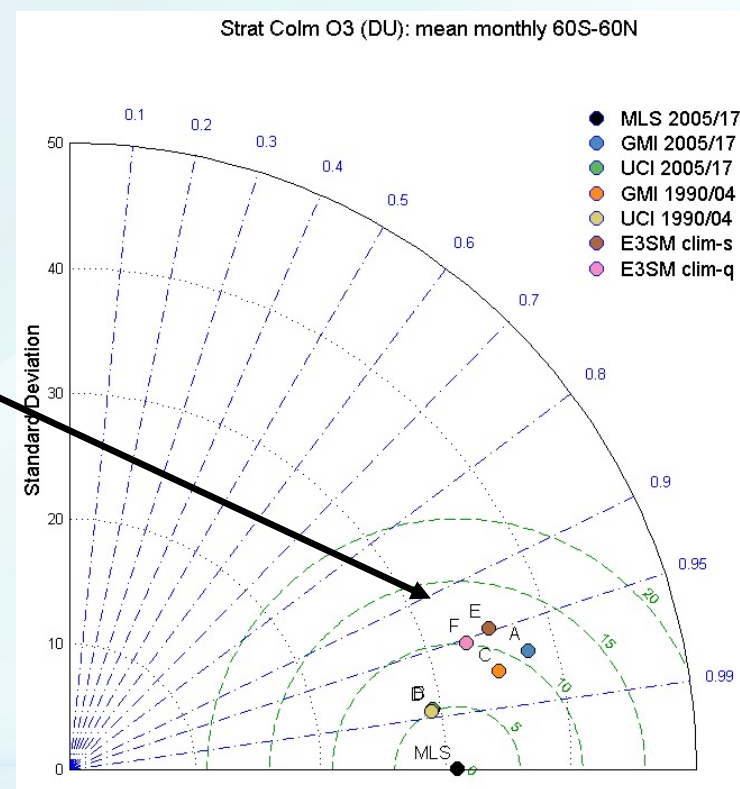
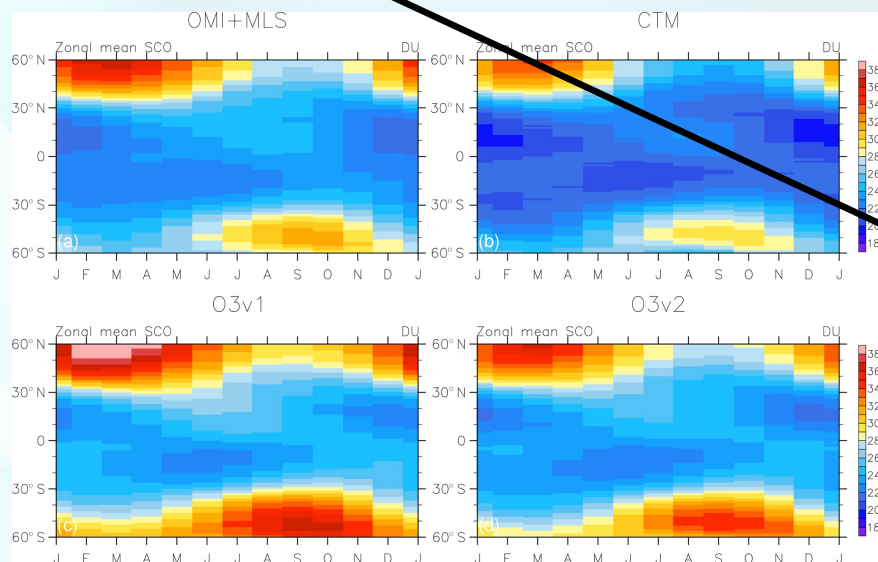


Fig. 6 | Ecosystem–atmosphere interactions exacerbate climate penalty on ozone extremes. **a–d**, The top row of map

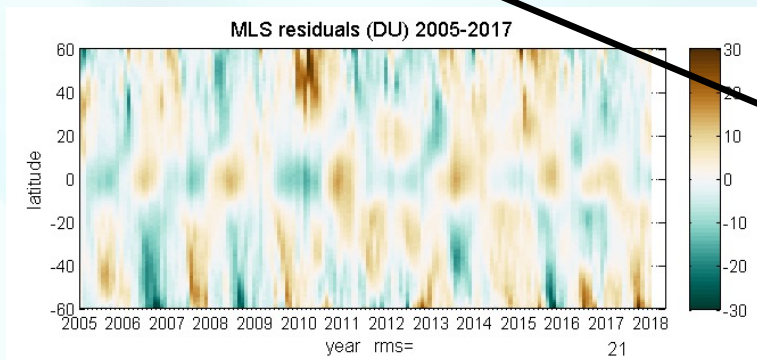
Stratospheric Column Ozone (SCO):

E3SM + Linoz v2 matches observations as well as UCI CTM and GSFC GMI CTM.

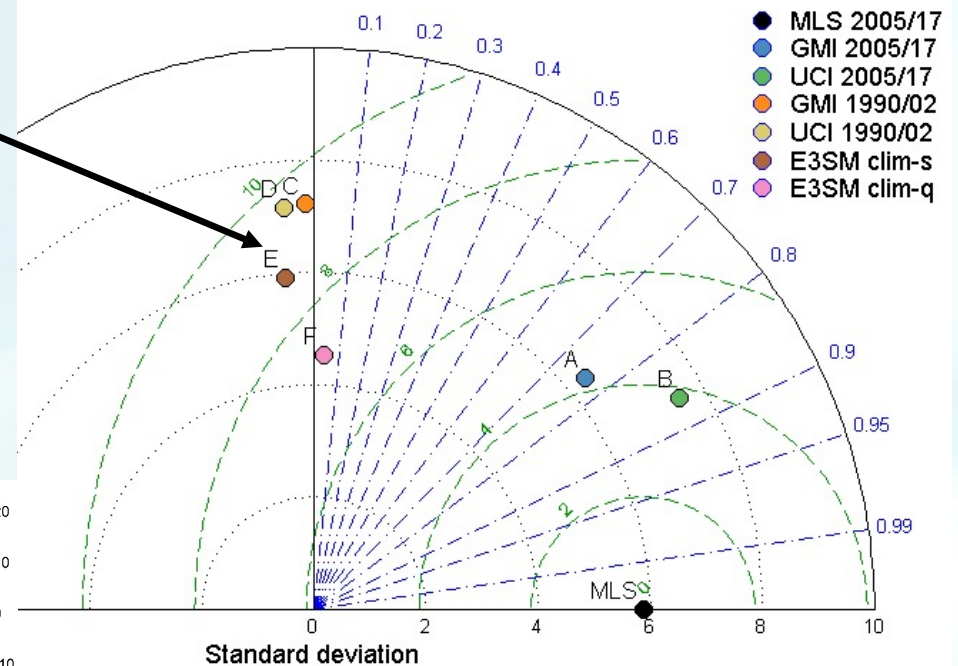


Stratospheric Column Ozone (SCO):

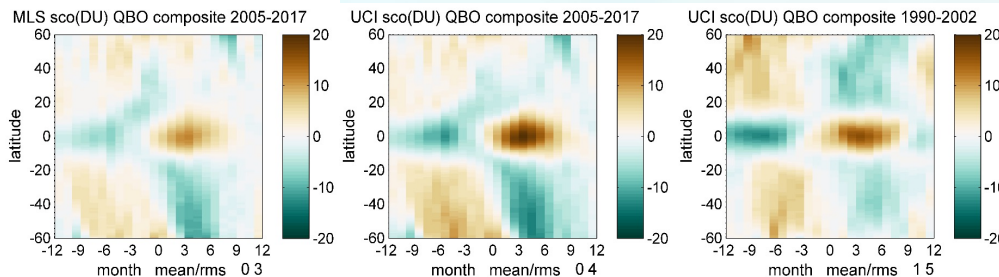
E3SM + Linoz v2 (climate model) **cannot** match interannual as well as UCI CTM and GSFC GMI. It fails in the same way as UCI and GMI do when sampled for different decade



SCO 13-Year Monthly Residuals 60S-60N (DU)

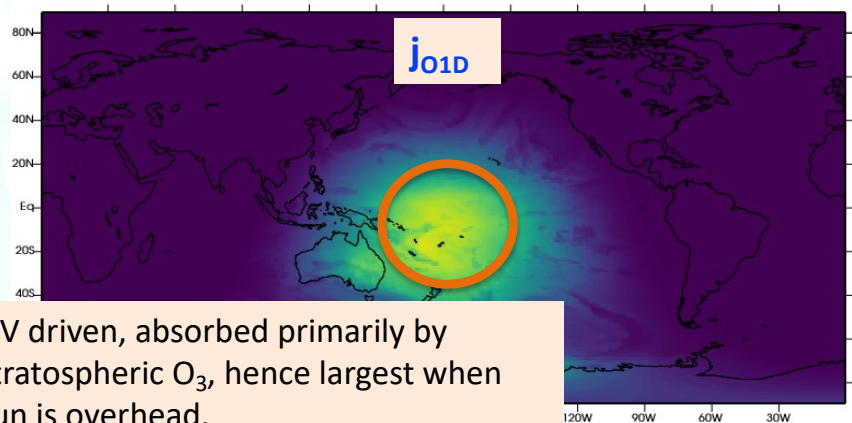
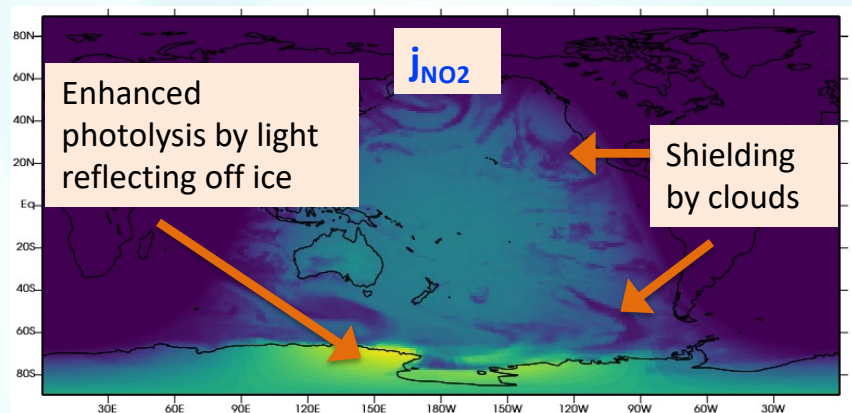


We designed a novel QBO composite that can be used for chemistry-climate models!



Fast-J implemented and coupled to photochemistry

Lat-lon maps January 2 00:00Z, Instantaneous



Fast-J is a huge improvement over lookup table.

- Removes lookup table biases in E3SM (Superfast chemistry)
- Consistent treatment with options for overlapping clouds
- Aerosol absorption and scattering
- Spherical geometry of atmosphere (Prather & Hsu, 2019)
- Multi-angle scattering:
 - Enhanced photolysis above clouds and in top of clouds.
 - Realistic diffuse **PAR incident** on ocean and biosphere
- Updated and updatable laboratory data tables
- Supported for global community by UC Irvine

Linoz v3: Linearized ozone chemistry for the stratosphere: the $\{N_2O, NO_Y, O_3, CH_4, H_2O\}$ -system

Four prognostic equations (dX/dt) for N_2O , NO_Y , O_3 and CH_4 , each a first-order Taylor expansion around monthly zonal mean chemical climatologies of overhead ozone column (photolysis rates) and local values of O_3 , N_2O , NO_Y , CH_4 , and temperature. Derivative terms are calculated with UCI's photochemical box (PRATMO).

Stratospheric H_2O is diagnosed from the CH_4 change, assuming a lower boundary (tropopause) value and conservation of potential water ($H_2O + 2xCH_4 = \text{constant}$).

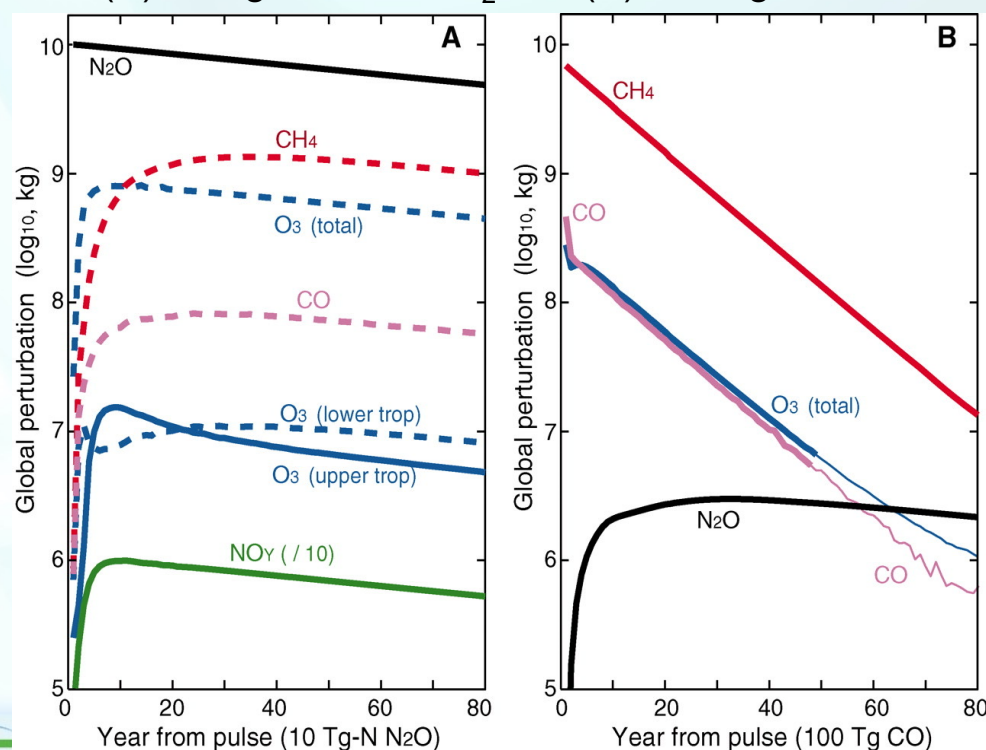
Stratospheric chlorine and bromine levels (Cl_Y , Br_Y) are set to a chemical climatology for the specified time period. The Linoz tables are calculated for the historical or projected levels of tropospheric halocarbons.

In contrast Linoz v2: O_3 only, one prognostic equation, linearized relative to climatology of overhead ozone column, O_3 and temperature. Tables are calculated for assumed tropospheric levels of halocarbons, CH_4 , and N_2O .

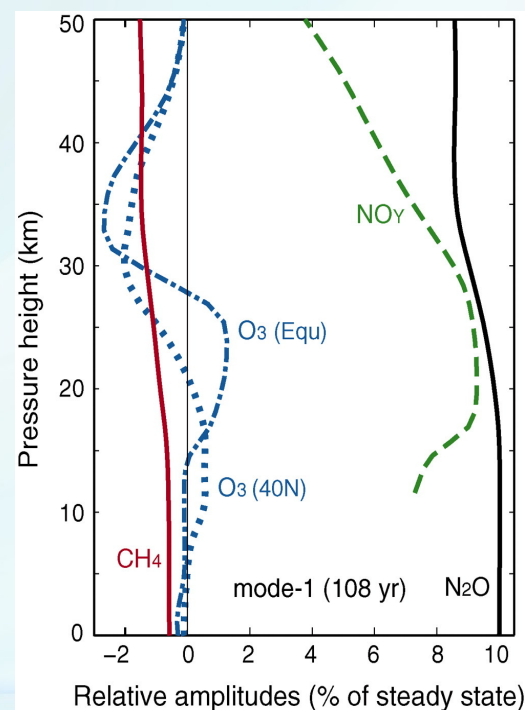
Using Linoz v3 with UCI chemistry CTM, we have the first identification of the link between the coupled N_2O - O_3 - NO_y stratospheric chemistry and CH_4 tropospheric chemistry through a 108-year global eigenmode.

Decay of atmospheric chemical perturbations caused by an initial pulse of

(A) 10 Tg of N from N_2O (B) 100 Tg of CO



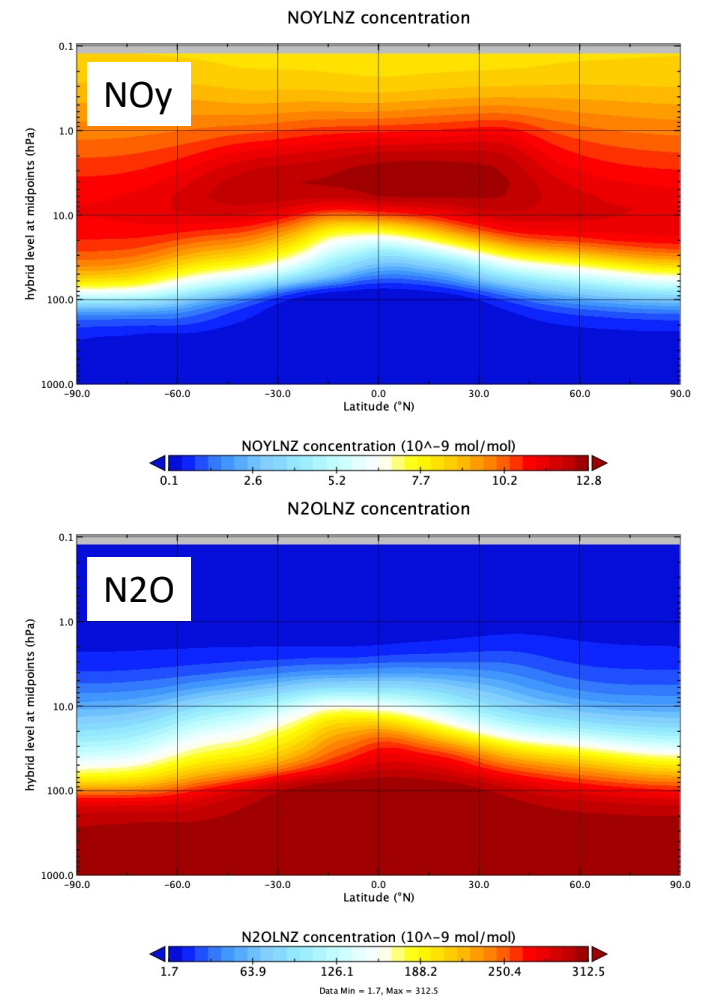
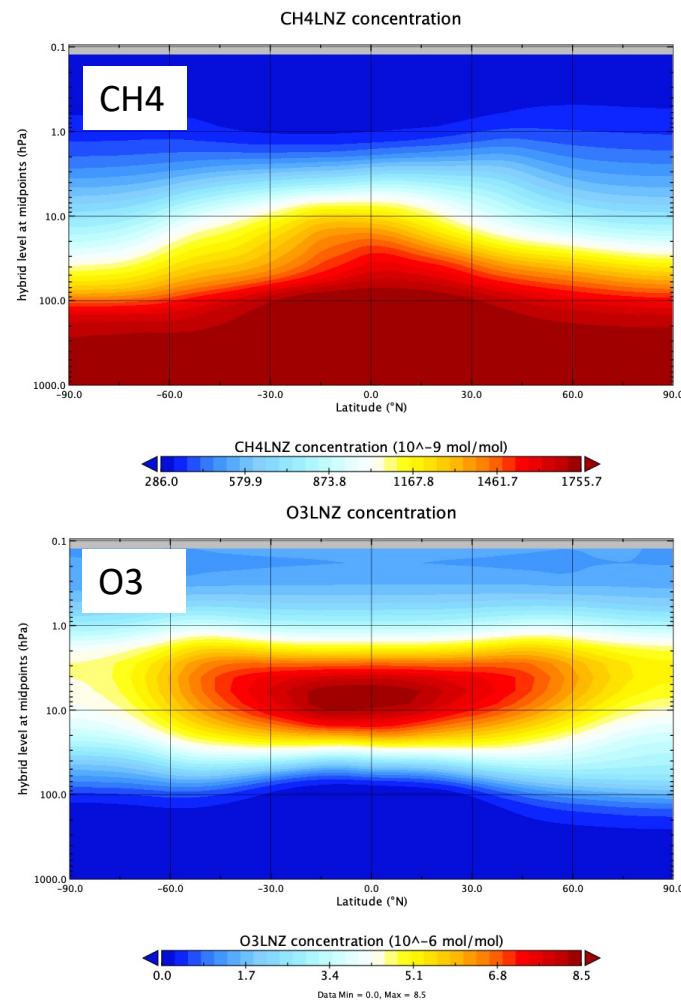
(C) Vertical profiles of Mode-1 in (A)



Prather and Hsu (2010) *Science*, 330:952-954.

Annual zonal mean of Linoz v3 species in an E3SM run are a close match to observations.

Already on the E3SM
GitHub branch

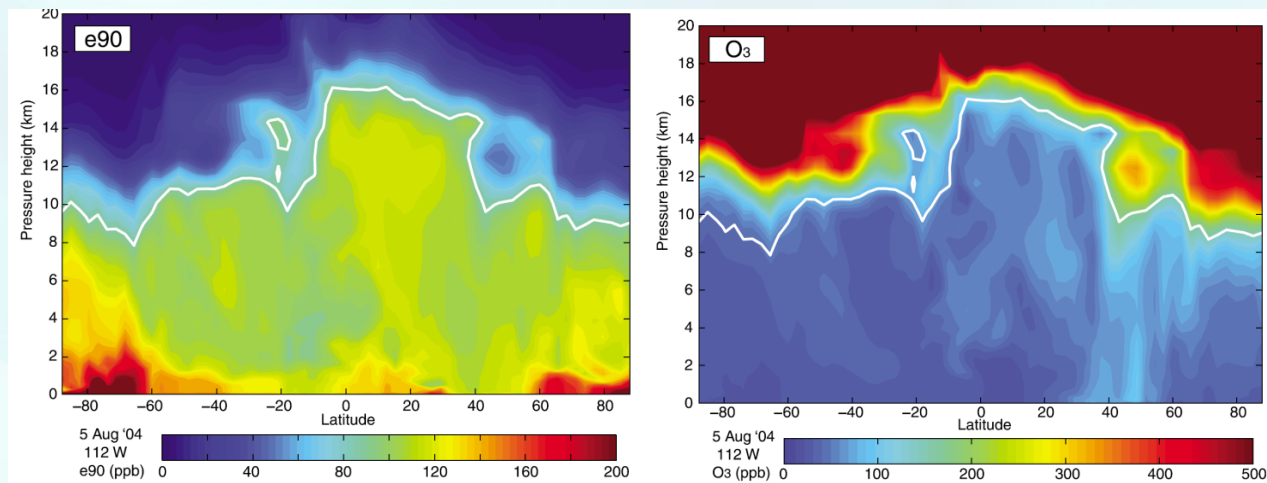


E90 tropopause clearly separates strat vs. trop in 3D

An atmospheric chemist in search of the tropopause

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 116, D04306, doi:10.1029/2010JD014939, 2011

Michael J. Prather,¹ Xin Zhu,¹ Qi Tang,¹ Juno Hsu,¹ and Jessica L. Neu^{1,2}



- With equivalent emissions, E3SM TPP threshold (75.5 ppb) is smaller than the UCI CTM threshold (89.8 ppb).
- A new diagnostic, suggesting **slower tropospheric mixing** in E3SM than in the EC winds
- Currently defined as a 2D TPP. Need to implement a 3D TPP in E3SM for chemistry

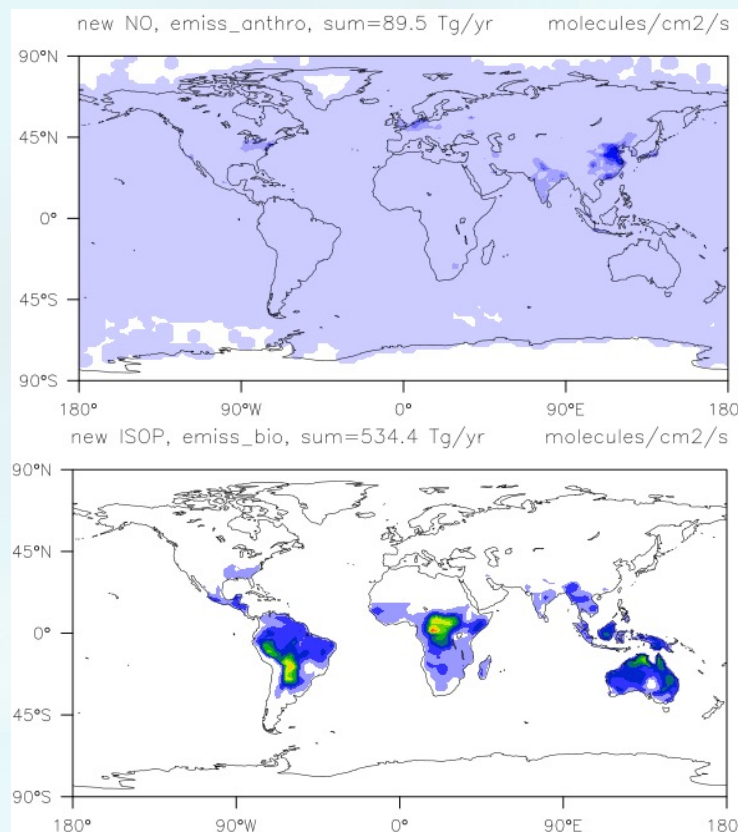
chemUCI (years 2000-2009)

CMIP6 emissions plus natural emissions

Year 2009

NO
sum = 89.5 Tg/yr

Biogenic Isoprene
sum = 534.4 Tg/yr

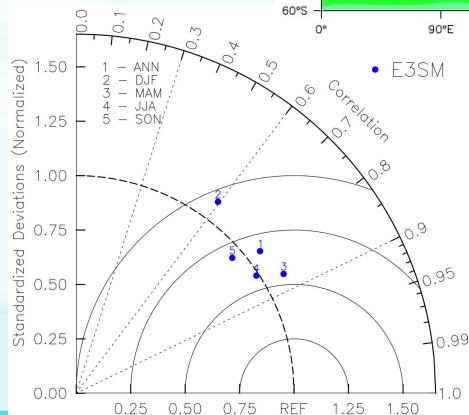
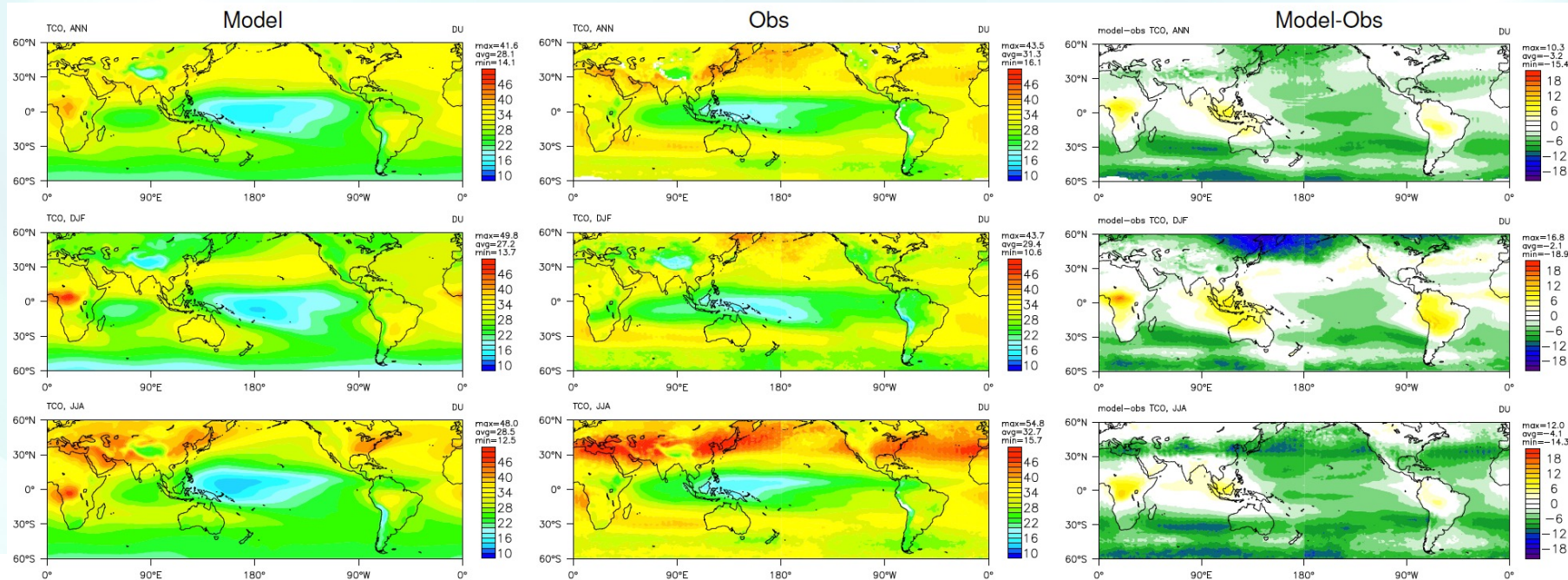


Trop column ozone vs. satellite obs (OMI/MLS)

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DJF

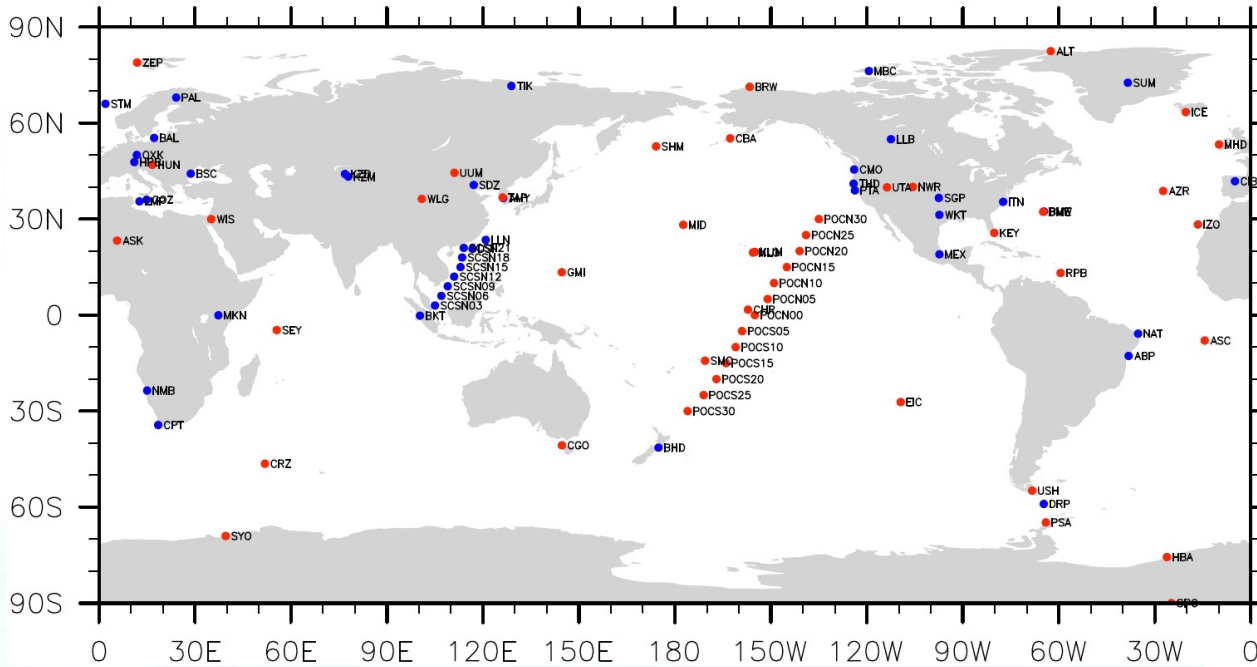
JJA



- Model overestimates over tropical land, while underestimates over oceans.
- Normalized root-mean-square difference ratios (0.9-1.2) and correlations (0.6-0.9) are reasonably captured.

Surface CO vs. NOAA obs

NOAA CO sites

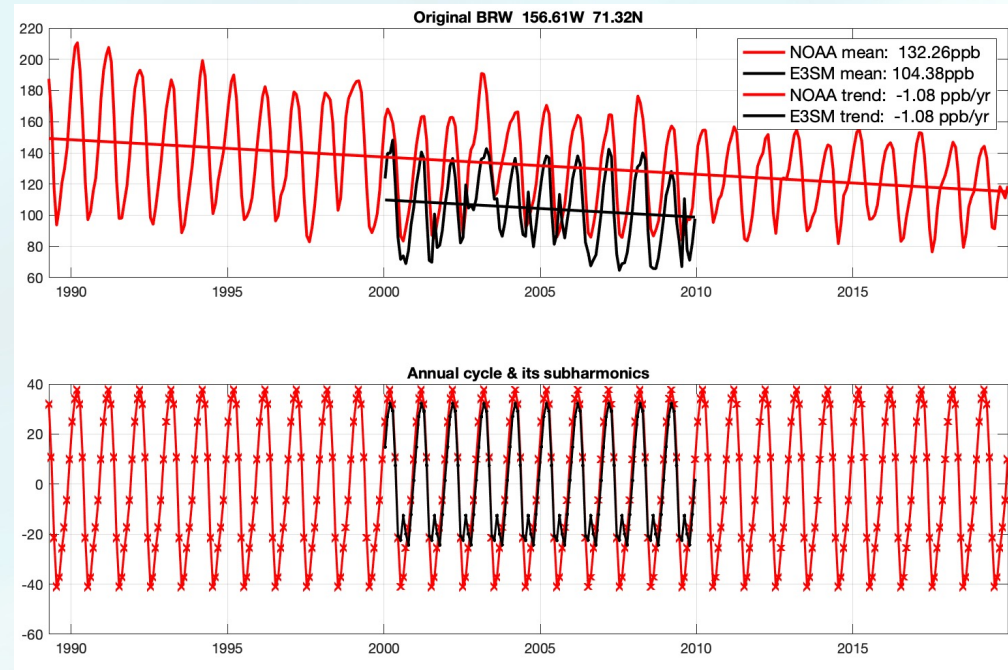


- We pick sites with long-time data (> 20 years)
- And not close to mountains and pollution



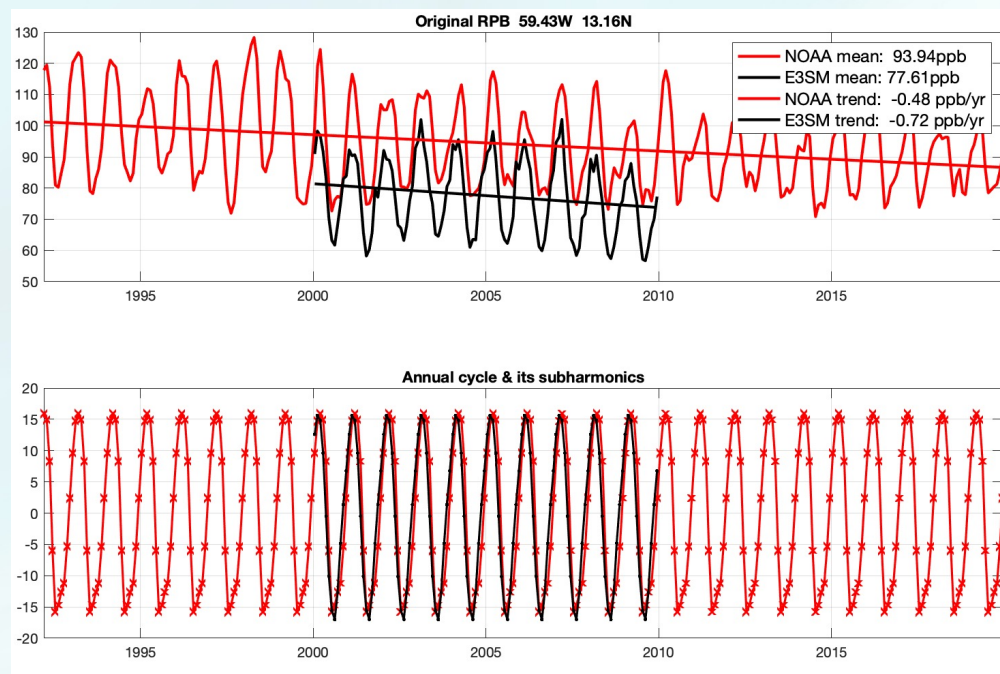
Surface CO vs. NOAA obs

BRW, 71.32 N, 156.61 W



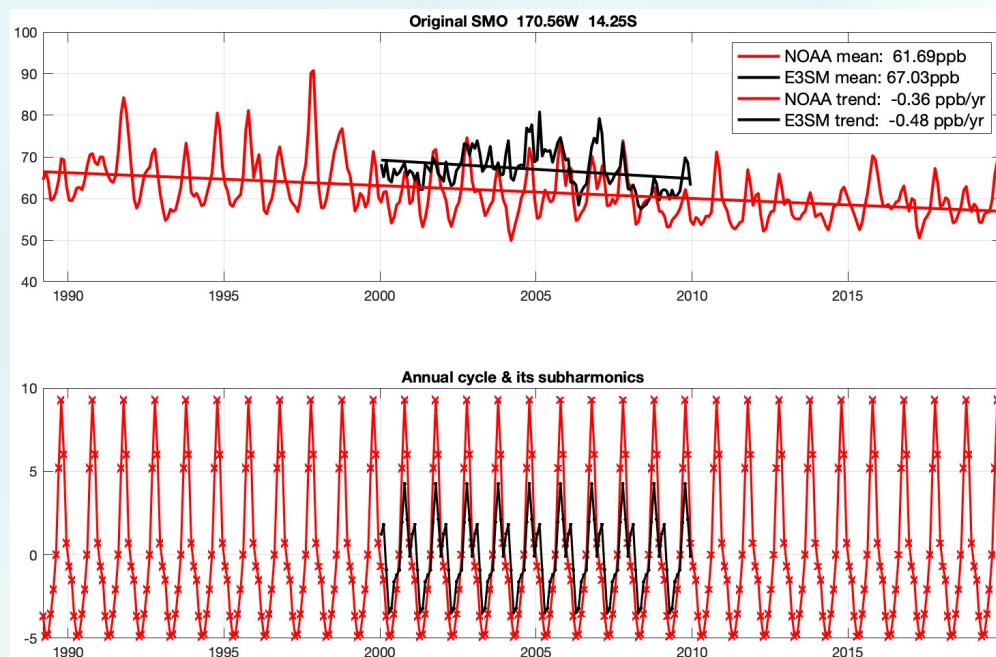
Surface CO vs. NOAA obs

RPB, 13.16 N, 59.43 W



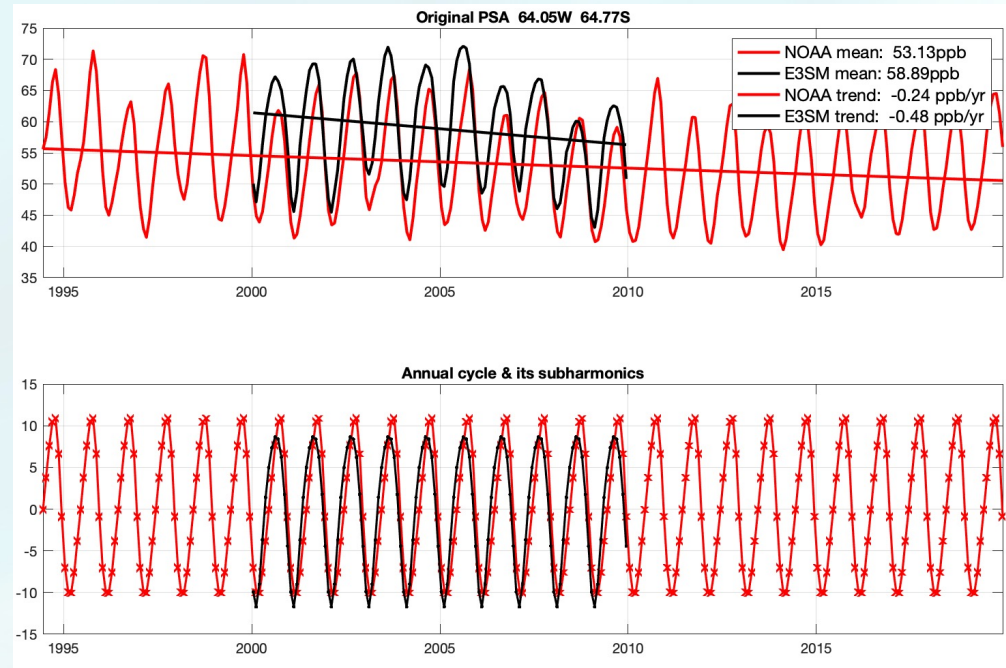
Surface CO vs. NOAA obs

SMO, 14.25 S, 170.56 W
ITCZ problems



Surface CO vs. NOAA obs

PSA, 64.77 S, 64.05 W

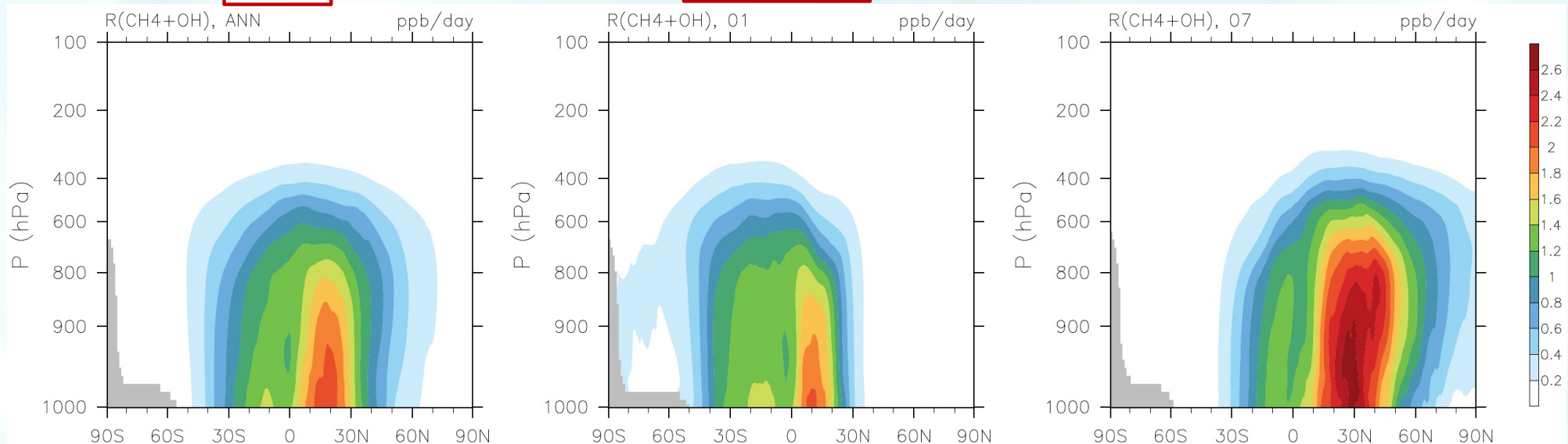


Methane loss rate (year 2000)

ANN

January

July



- Methane is prescribed in this run.
- Changes in the methane loss rate imply OH changes.

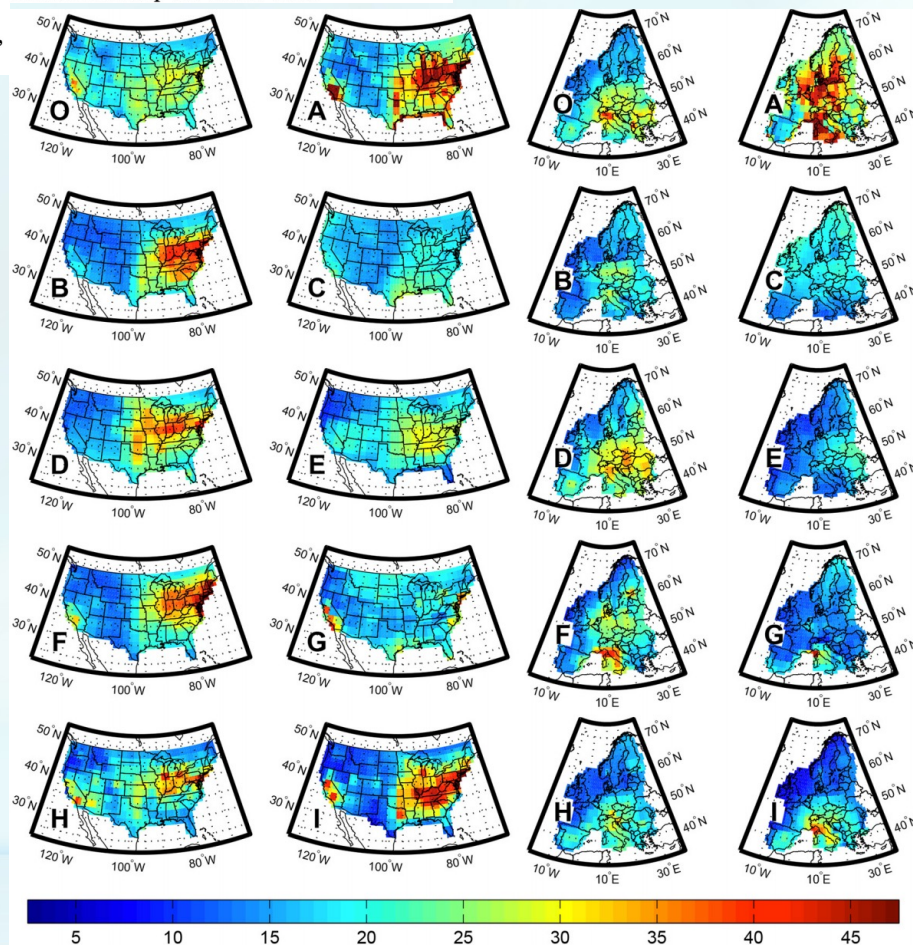
More metrics – air quality (O₃ and PM)

Use of North American and European air quality networks to evaluate global chemistry–climate modeling of surface ozone

J. L. Schnell¹, M. J. Prather¹, B. Josse², V. Naik³, L. W. Horowitz⁴, P. Cameron-Smith⁵, D. Bergmann⁵, G. Zeng⁶, D. A. Plummer⁷, K. Sudo^{8,9}, T. Nagashima¹⁰, D. T. Shindell¹¹, G. Faluvegi¹², and S. A. Strode^{13,14}

Atmos. Chem. Phys., 15, 10581–10596, 2015
www.atmos-chem-phys.net/15/10581/2015/
doi:10.5194/acp-15-10581-2015

Use air quality networks
observations to evaluate
modeled surface O₃



More metrics - ATom

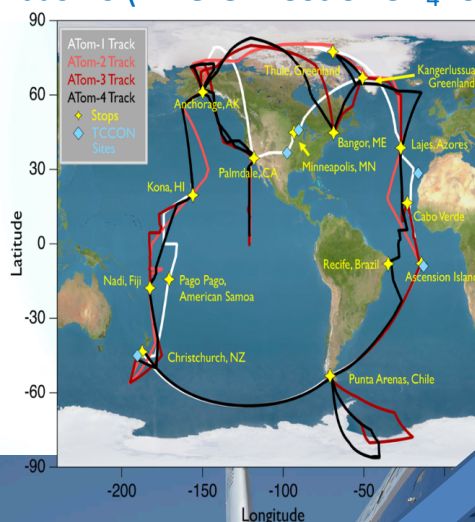
NASA Atmospheric Tomography Mission, 2016-2021

Review status: this preprint is currently under review for the journal ACP.

Heterogeneity and Chemical Reactivity of the Remote Troposphere defined by Aircraft Measurements

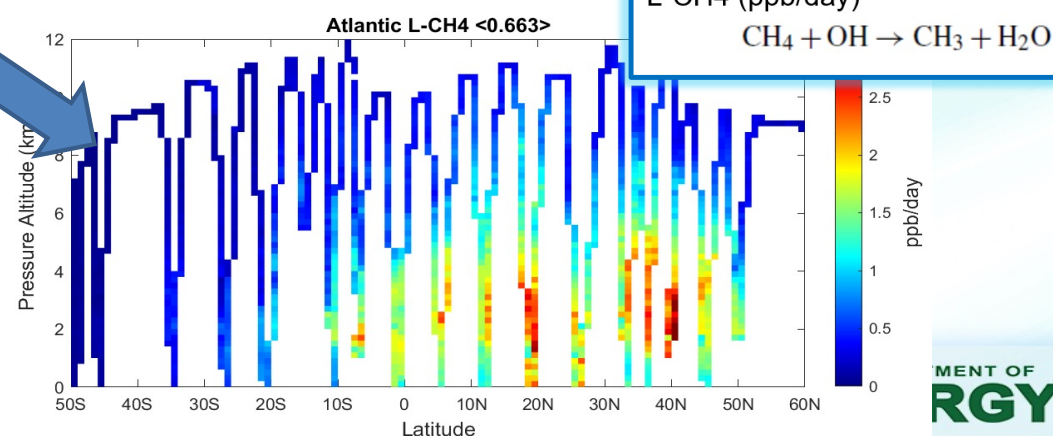
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Global profiling transects of the ocean basins (where most of CH₄ loss occurs)



Species

O₃, NO_x
CO
HNO₃, CH₃OOH, HNO₄, H₂O₂
HCHO
CH₄
PAN
MeONO₂
EtONO₂ and higher organic nitrates
Isoprene
Acetone, Acetaldehyde
Methanol
Ethane and higher alkanes
Ethene, Ethyne



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Summary

- We implemented chemUCI + Linoz v3 interactive chemistry in an E3SMv2 beta code.
 - Initial results show reasonable tropospheric column ozone and surface CO compared to observations
 - Ready for BGC and aerosol links
 - Need further evaluation against observations and other models
 - Define new chemistry metrics for E3SM diagnostics.
- Working with BGC group, define interface for interactive methane and ozone simulations.
- With aerosol team to link chemistry and aerosols