Modeling Atmospheric Chemistry and Aerosols

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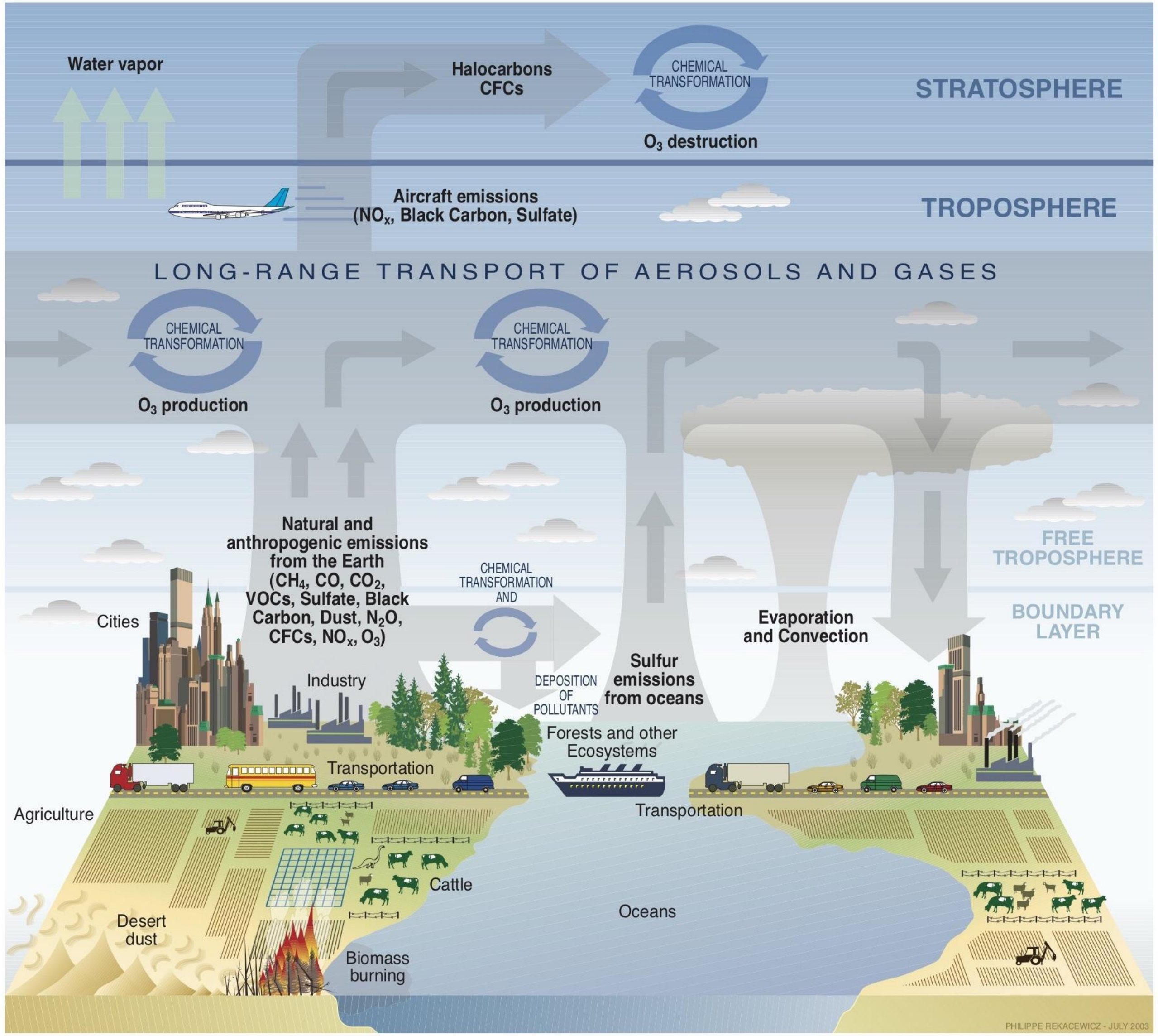
University of Wollongong, Australia → NSF NCAR

Atmospheric Chemistry

- Motivation
- Adding processes into models
  - Emissions
  - Chemical mechanism
  - Aerosol model and cloud interactions
  - Transport
  - Dry Deposition
  - Wet Deposition
- Applications
- Summary



Atmospheric Chemistry: Why it is important – Health

Tropospheric ozone pollution (NOx, CO, VOC, CH4):
→ Damages tissues, causes inflammation
→ Coughing, chest tightness and worsening of asthma

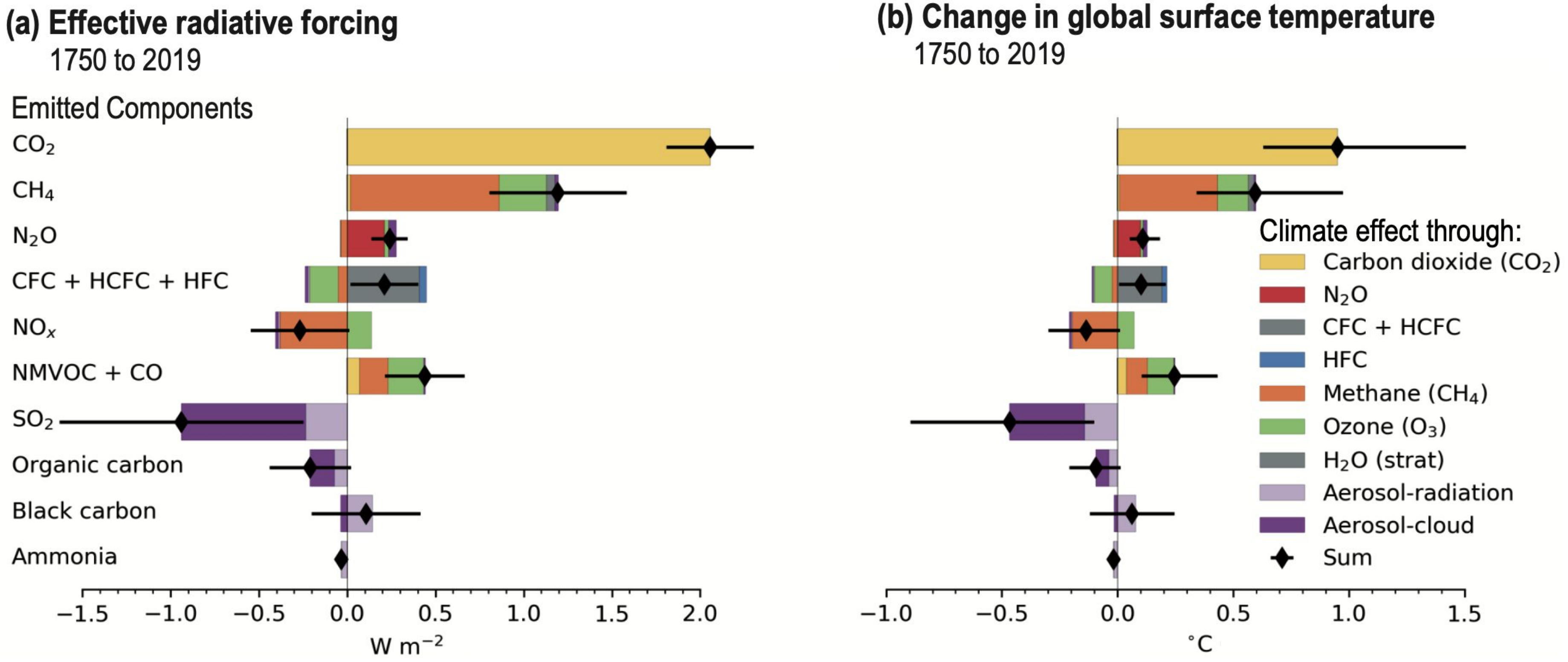
Particulate Matter: PM2.5 and PM10 diameter < 2.5 or 10 µm (SO2, VOC, NH3, BC, OC, fine dust):
→ Cardiovascular impacts (lungs and heart), premature deaths

Sources:

- Traffic / Industry & Private (use of fossil fuels)
- Farmland
- Fires
- Vegetation
- PM: Dust storms (worsen with climate change)
- PM: Volcanoes

(7+ million premature deaths due to air pollution per year !!)

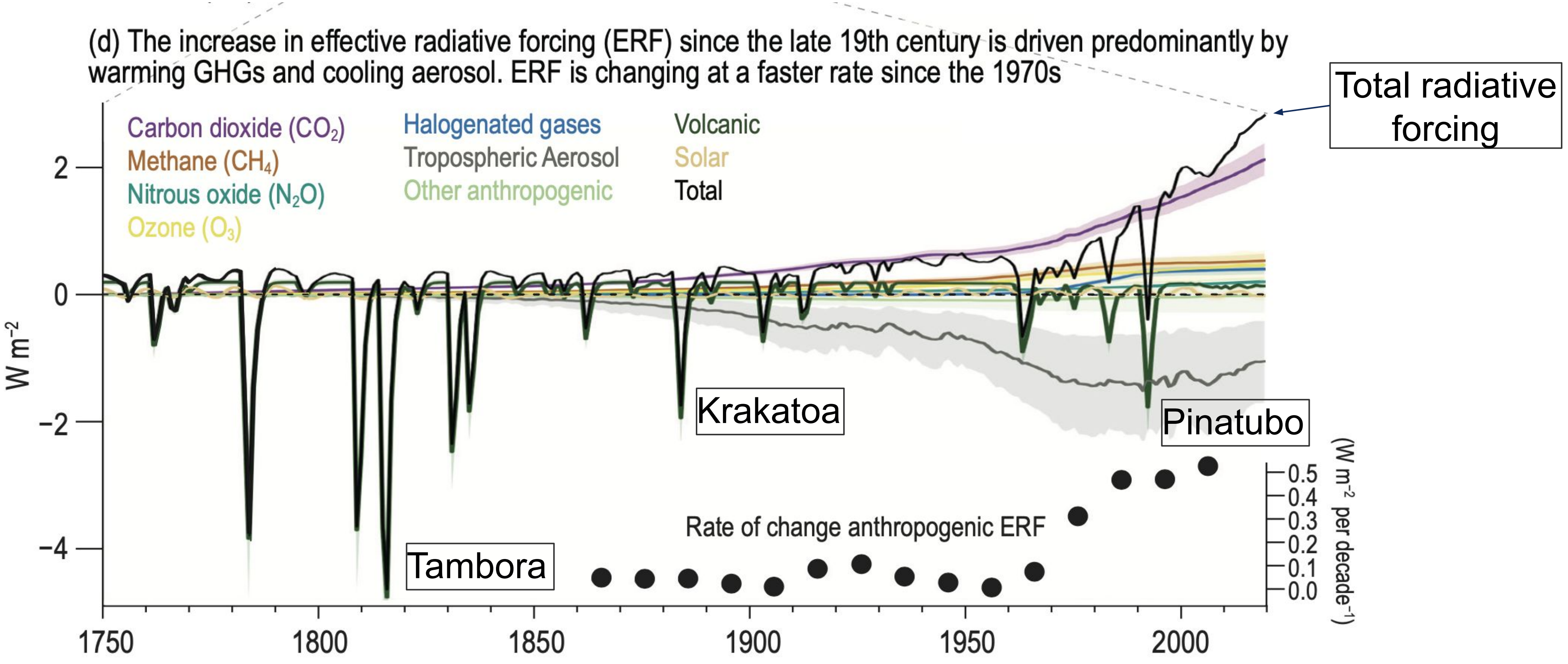
Atmospheric Chemistry: Why it is important – Climate



IPCC AR6 WG1 Technical Report, Figure TS15

- Chemistry and aerosols interact with the climate
- Importance of describing ozone and aerosol precursors
- Importance of aerosol-cloud interactions in models

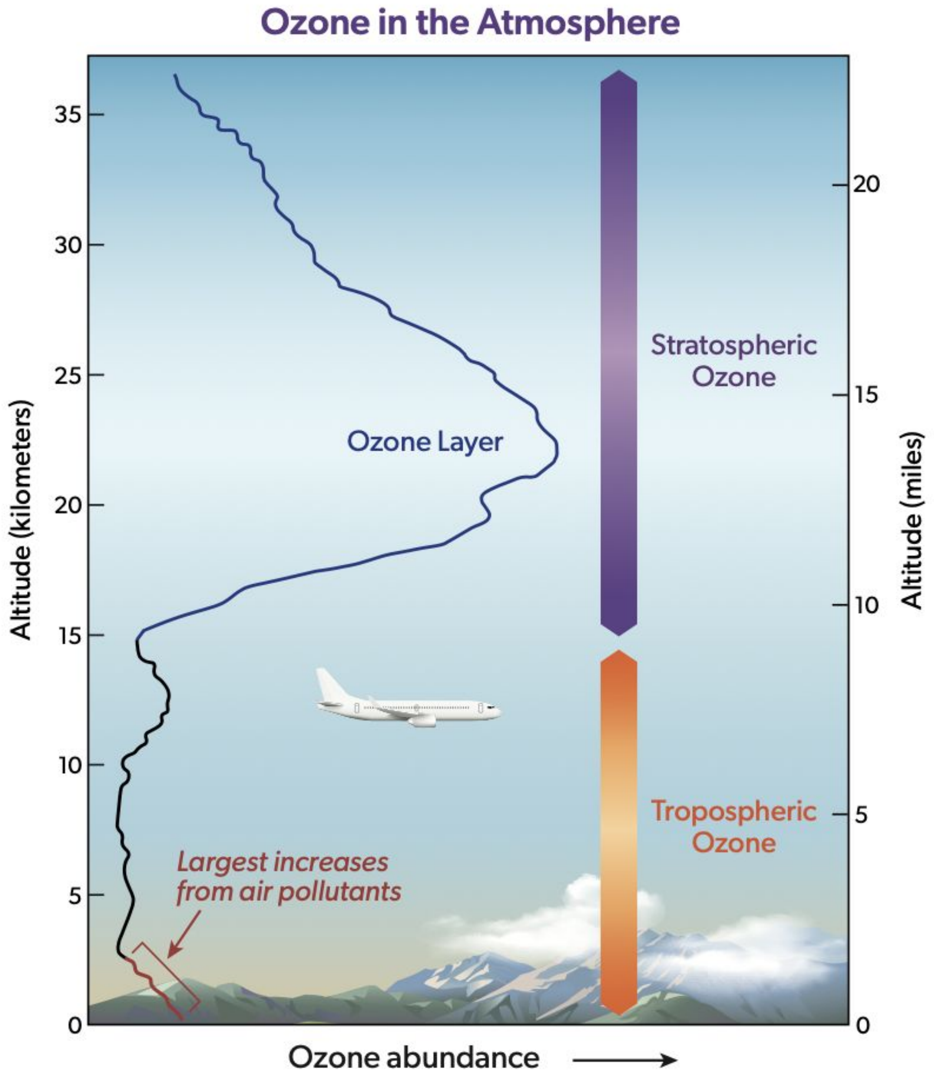
Atmospheric Chemistry: Why it is important – Climate

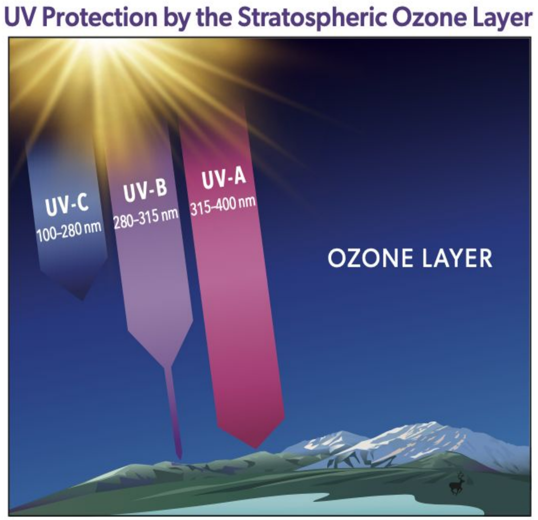


IPCC AR6 WG1 Technical Report, Figure TS9

Chemistry and aerosols interact with the climate system,
-> need to be well described in climate models

Atmospheric Chemistry: Why is it important – Stratospheric Ozone





The ozone layer in the stratosphere protects life from harmful UV, through photochemical reactions

Accurate modeling is required:

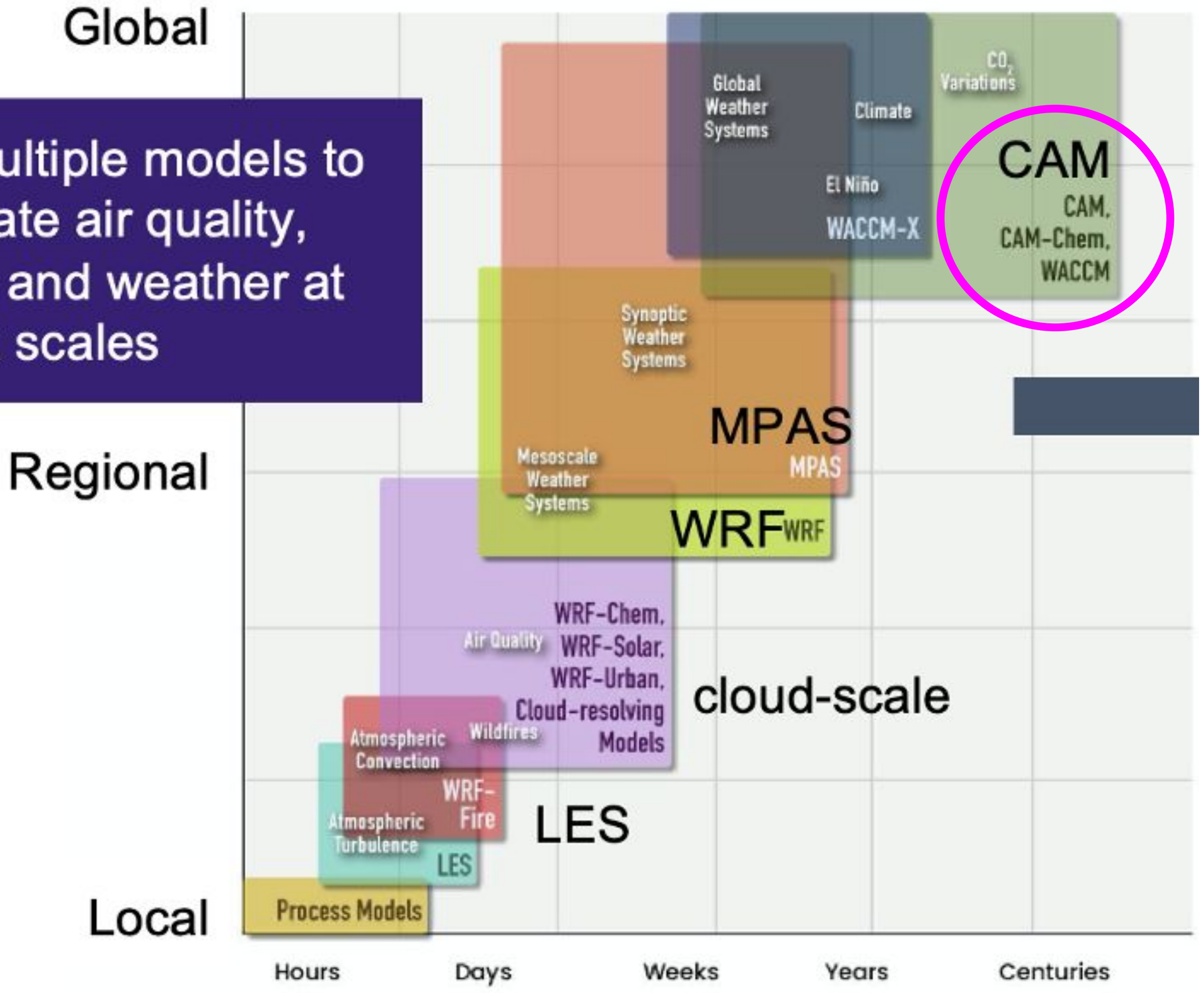
- Impact on tropospheric chemistry
- Ozone hole recovery (CFCs)
- Cause of a slowing trend

Different atmospheric models

Many different types of atmospheric models: e.g. Box, Column, Large Eddy Simulations, Limited Region, Global, Chemical Transport Models, Earth System Models

Models include approximations: it is important to **use the best tool for your question**. There are often different options, even within one “type” of model, such as the Community Earth System Model (CESM).

Need multiple models to investigate air quality, climate, and weather at different scales

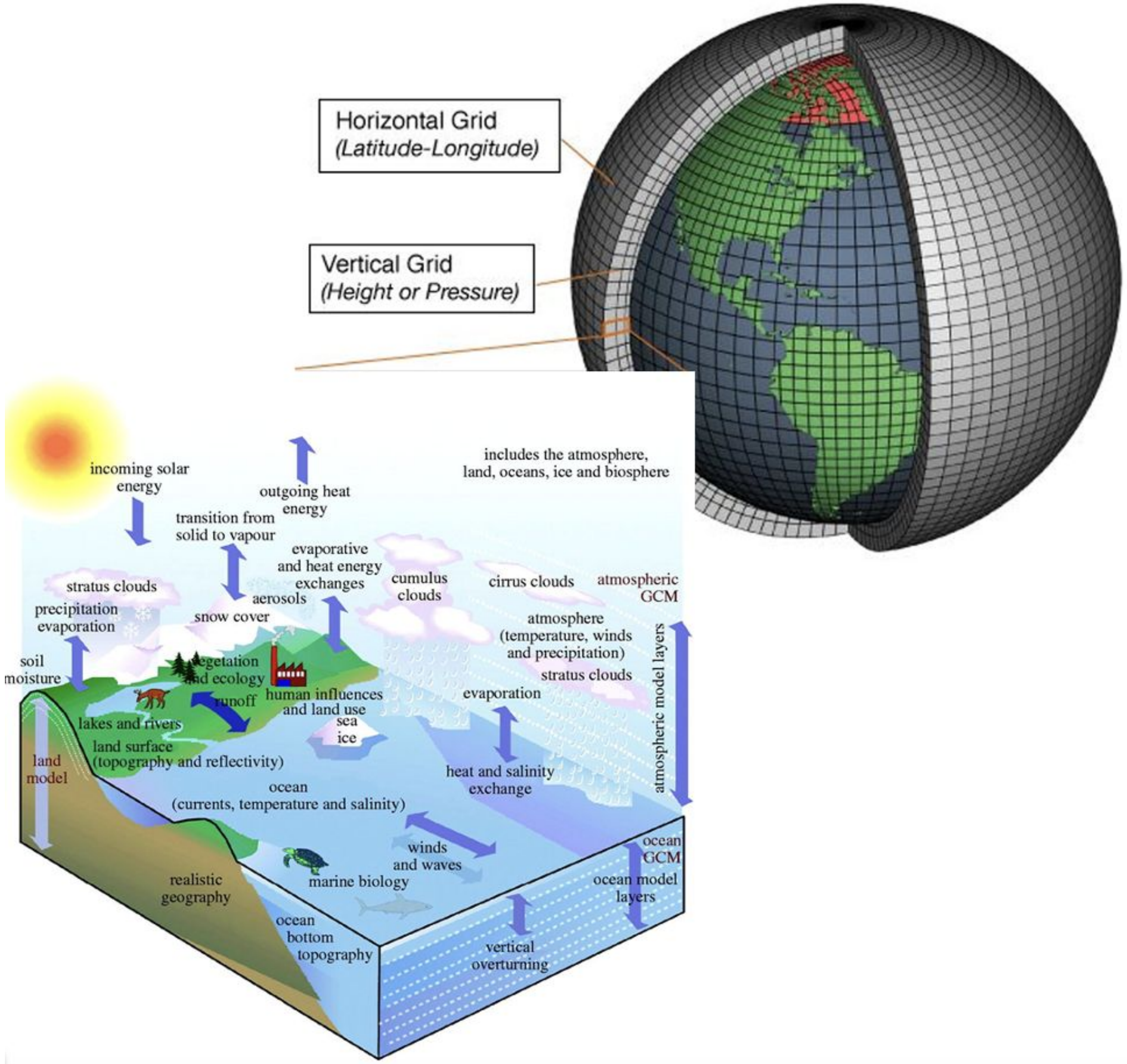


Global Earth System Models

These models use physical equations to simulate key fields and processes in the atmosphere, ocean, land, sea-ice, land-ice, etc.

Processes that remain below the grid resolution need to be parameterized.

ESMs build on our understanding of processes from observations and highly-detailed models (e.g., process models, large eddy simulations).



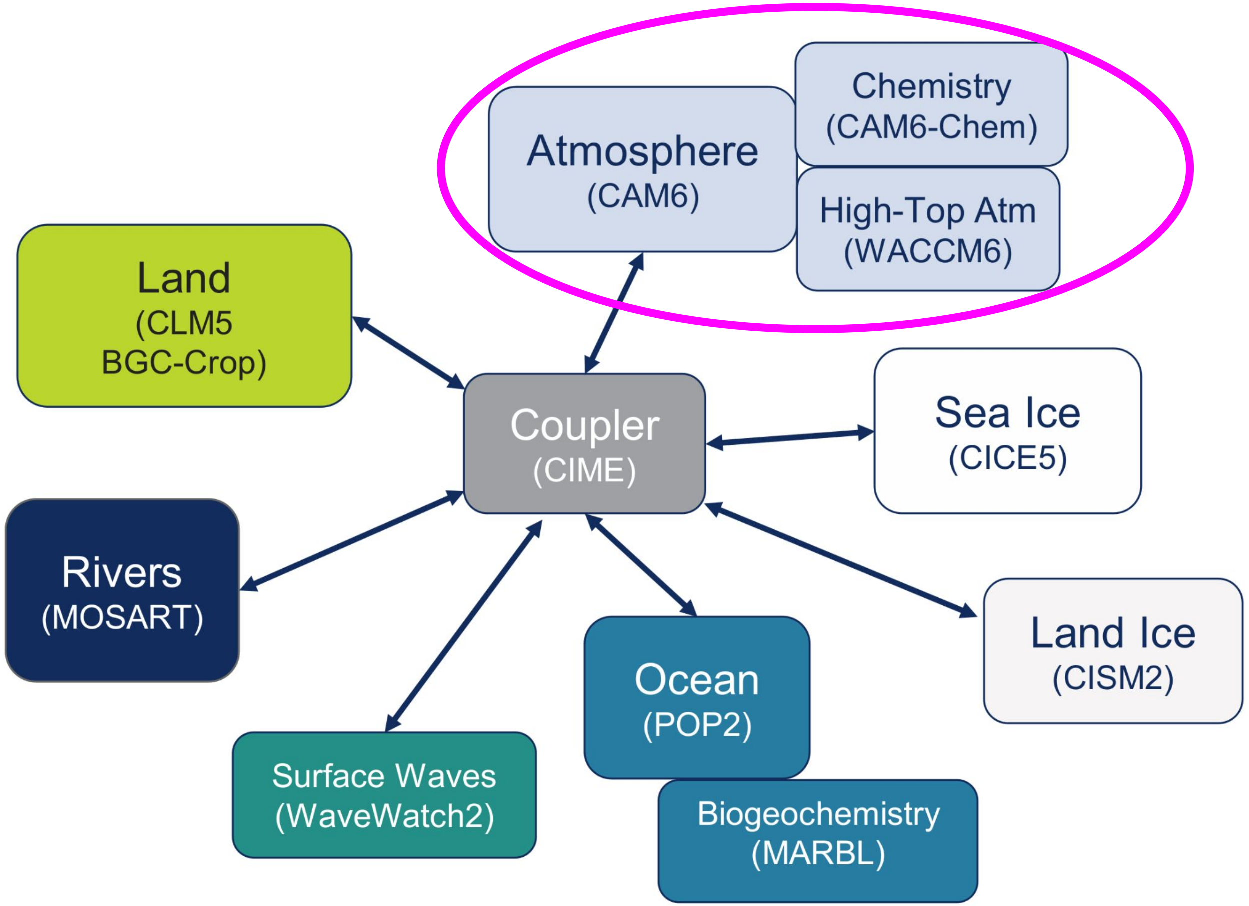
The Community Earth System Model (CESM)

CESM has multiple different earth system components coupled with a coupler.

The atmosphere model in CESM is called **CAM** (Community Atmosphere Model)

The Community Atmosphere Model with Chemistry (**CAM-chem**) is a component of CESM.

When running CAM-chem, the land component is on by default. Other components can be on or off using different settings.



Atmospheric Chemistry

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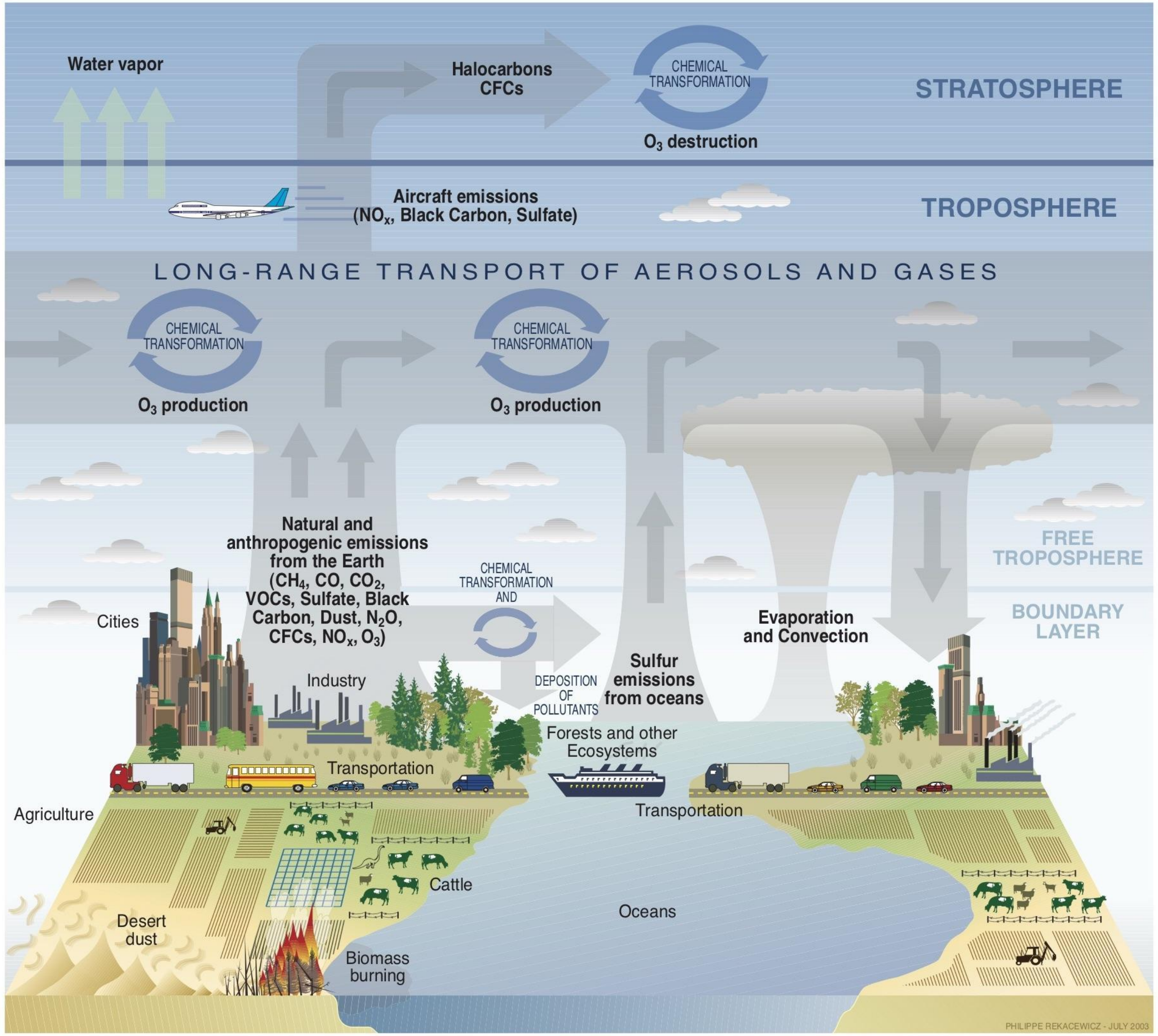

For each chemical constituent (χ), the following must be solved

$$\frac{\partial \chi(i)}{\partial t} = \mathrm{Sources(i)} - \mathrm{Sinks(i)}$$

Introduction to Atmospheric Chemistry, Daniel J. Jacob https://acmg.seas.harvard.edu/education/introduction-atmospheric-chemistry

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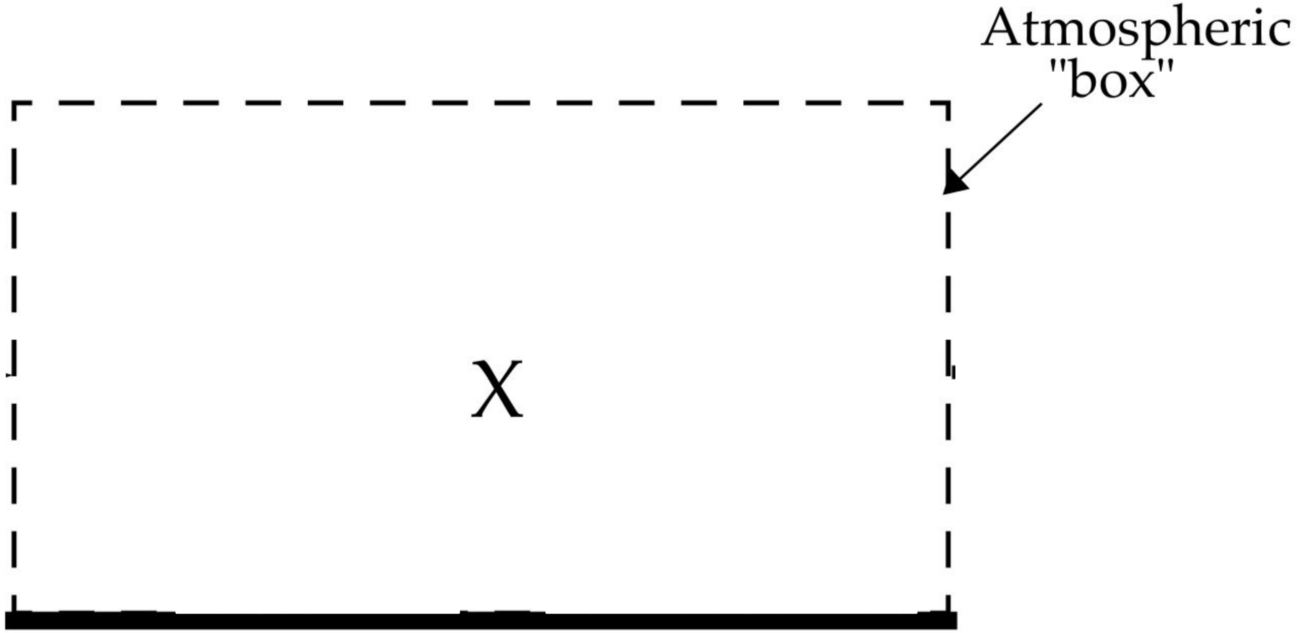

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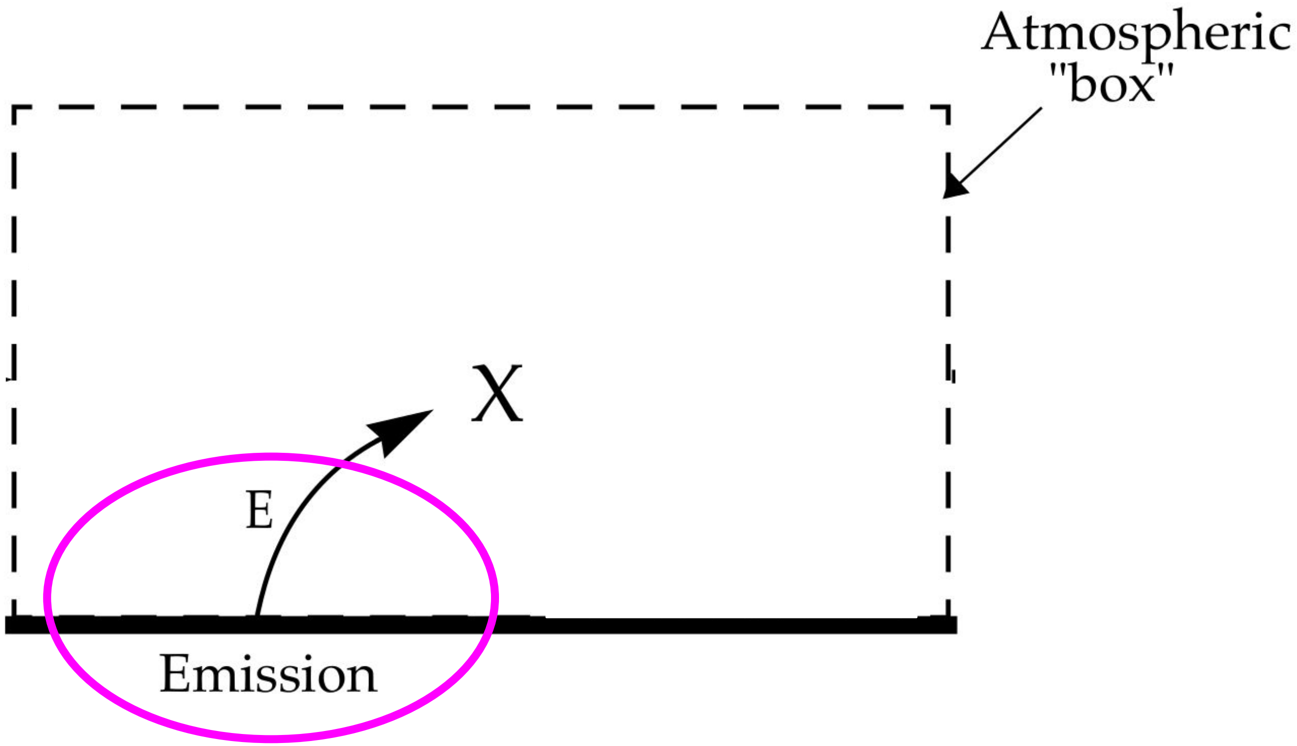
$$\frac{\partial \chi(i)}{\partial t} = \mathrm{Sources(i)} - \mathrm{Sinks(i)} = \mathrm{E_i} + \mathrm{C_i} + \mathrm{A_i} + \mathrm{T_i} - \mathrm{W_i} - \mathrm{D_i}$$

E_i Emissions



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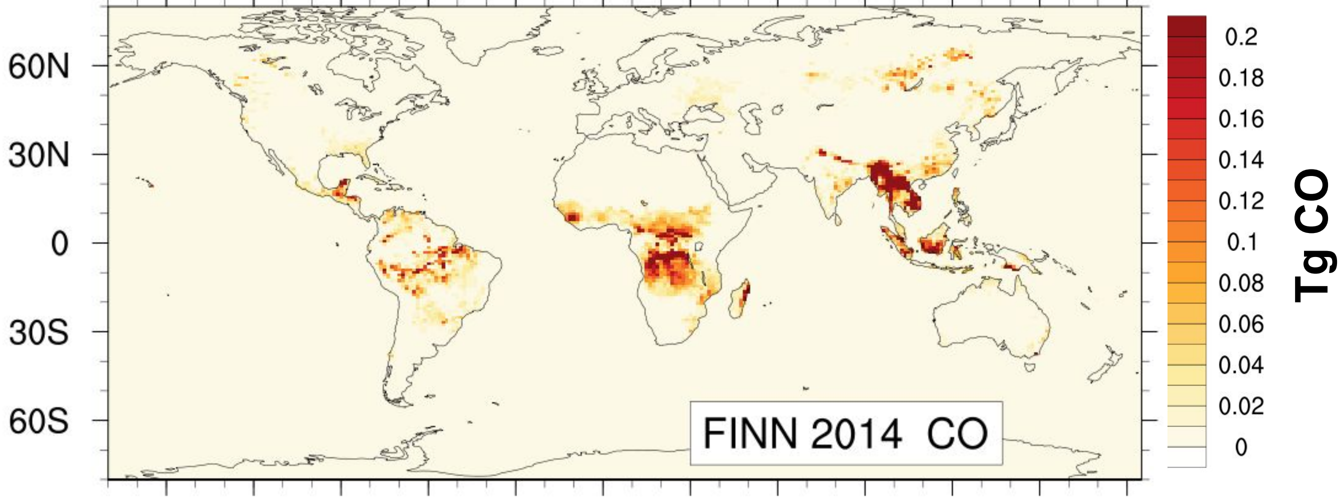
Emissions in CESM: 4 main “types”

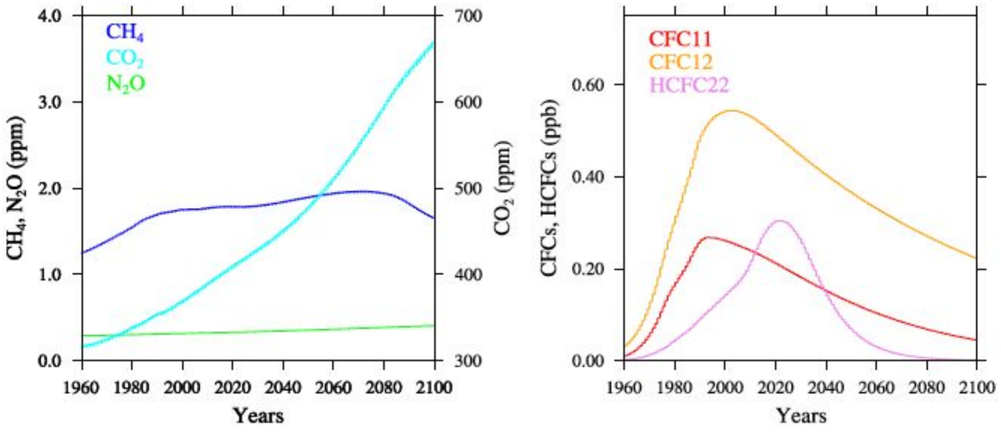
Emissions

- Surface emissions: anthropogenic, biogenic, biomass burning (fire), ocean, soil
- Vertical emissions: (external forcings): aircraft, volcanoes, power plants, (fire optional)
- Interactive: Dust, biogenic, sea salt, lightning NO_X, (fire optional/experimental)

Surface concentrations

- Lower boundary conditions (greenhouse gases CO_2, CH_4, O_3, N_2O and, long-lived gases CFCs). Can vary latitudinally.


Fire emissions of CO


Lower Boundary Conditions, RCP6.0

Biomass Burning Emissions

- Smoke is a complicated mixture of chemicals, including both trace gases and aerosols
- Biomass burning emissions are generally specified with offline gridded emissions files.
- In CESM:
  - CMIP6 (1750-2015)
  - GFED
  - QFED (near-real-time, NRT, and historical)
  - FINNv2.5 (2002-2023, and NRT)
  - GFAS (in progress)

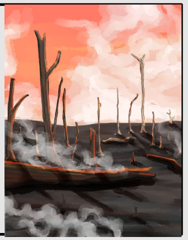

Image: Sam Hall, NSF NCAR

https://wiki.ucar.edu/display/camchem/Emission+Inventories

Creating biomass burning emission inventories

Method 1

Burned area
x
Biomass loading and fraction burned



GFED (CMIP6) FINN (active fire)

Fuel consumption
(kg C burned)

x

Biome-specific
Emission Factors
kg/kgC
From field and lab studies (e.g. Akagi et al. 2011)

→ Emissions

Method 2

Fire radiative power (FRP)
x
Biome-specific conversion factors

GFAS QFED

Artwork: Caparelli ArtNScience

Sources of uncertainty: biomass burning emissions

Emissions

Fire Detection and burned area

relies on MODIS* (instrument changes), miss smaller fires, overpass times, cloud interference

*20+ years of MODIS observations but also available from, e.g., VIIRS, Sentinel-3

Emission Factor

multiple uncertainties & variability: aggregation of biomes, instrument uncertainty

Biome/vegetation Type

aggregation and definition of biomes/land cover, peat is not always included, misidentification, estimation of fuel consumption

Combustion Stage

flaming versus smouldering is not represented, and is important for designating emissions factors and quantifying total emissions

Discussion Section 4: Pan et al., ACP., 2020

Other uncertainties in emissions

missing species; injection height

Emissions: Anthropogenic

Anthropogenic emissions are specified in offline gridded emissions files, developed using "bottom-up" methods. Current inventories include:

- CMIP6 (CEDS) (Hoesly,et al. GMD, 2018, https://gmd.copernicus.org/articles/11/369/2018/)
- CAMS (Copernicus Atmosphere Monitoring Service)
  (Granier et al, 2019, https://hal.science/hal-02322431/)

HEMCO (Harmonized Emissions Component) is available in CESM3(beta), allowing for:

- easy combination of regional inventories (NEI, etc.) with global inventories
- application of diurnal variation
- application of vertical distribution (power plant heights)

"Other" offline emissions

Climatological gridded inventories are used for soil and ocean emissions: Ocean CO and hydrocarbons, Soil NO, Soil NH_3

Interactive emissions: Dust

$F_{dustemis} = F_{dustemis}(u_*, w)$ becomes $F_{dustemis} = F_{dustemis}(u_*, w, \underbrace{z_{0,rock}, LAI}_{\text{Drag partition due to surface roughness}}, \sigma_{\tilde{u}})$

(CESM default)

horizontal wind/friction

soil moisture

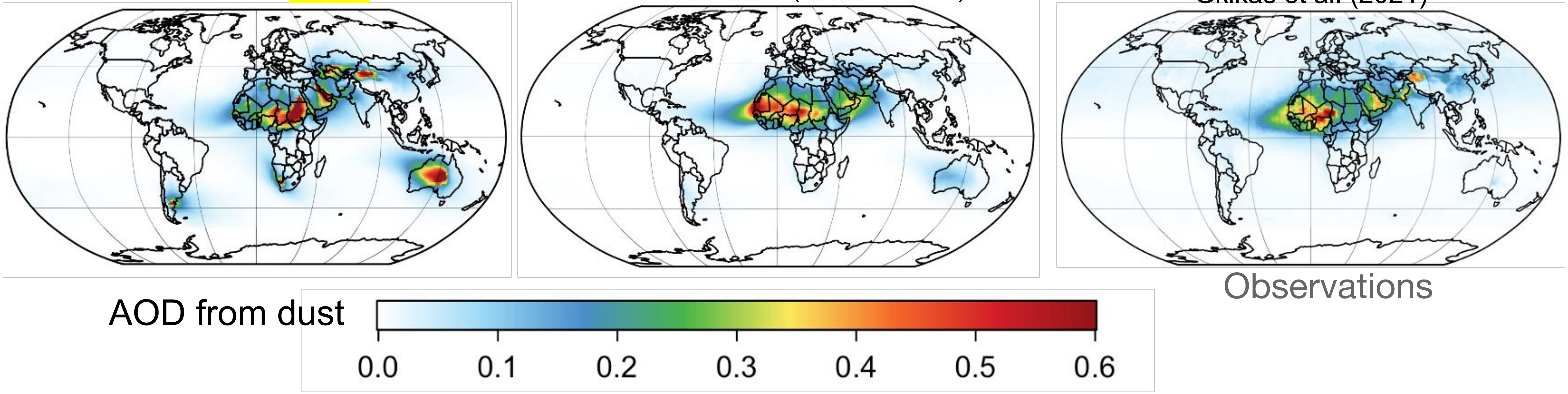
(Leung 2023)

Subtimestep wind following the similarity theory

Charlie Zender et al. (2003; DEAD)
CESM2/CAM6 default

Danny Leung et al. (2023; L23)
CESM3/CAM7 (future default)

MIDAS (MODIS/Aqua) dust
Gkikas et al. (2021)



Interactive emissions: Biogenic

MEGAN: Model of Emissions of Gases and Aerosols from Nature

A modeling system to estimate emissions of gases and aerosols from terrestrial ecosystems. The MEGANv2.1 algorithm is included in CESM within the Community Land Model (CLM) and uses model vegetation and meteorology.

Emissions for species i:

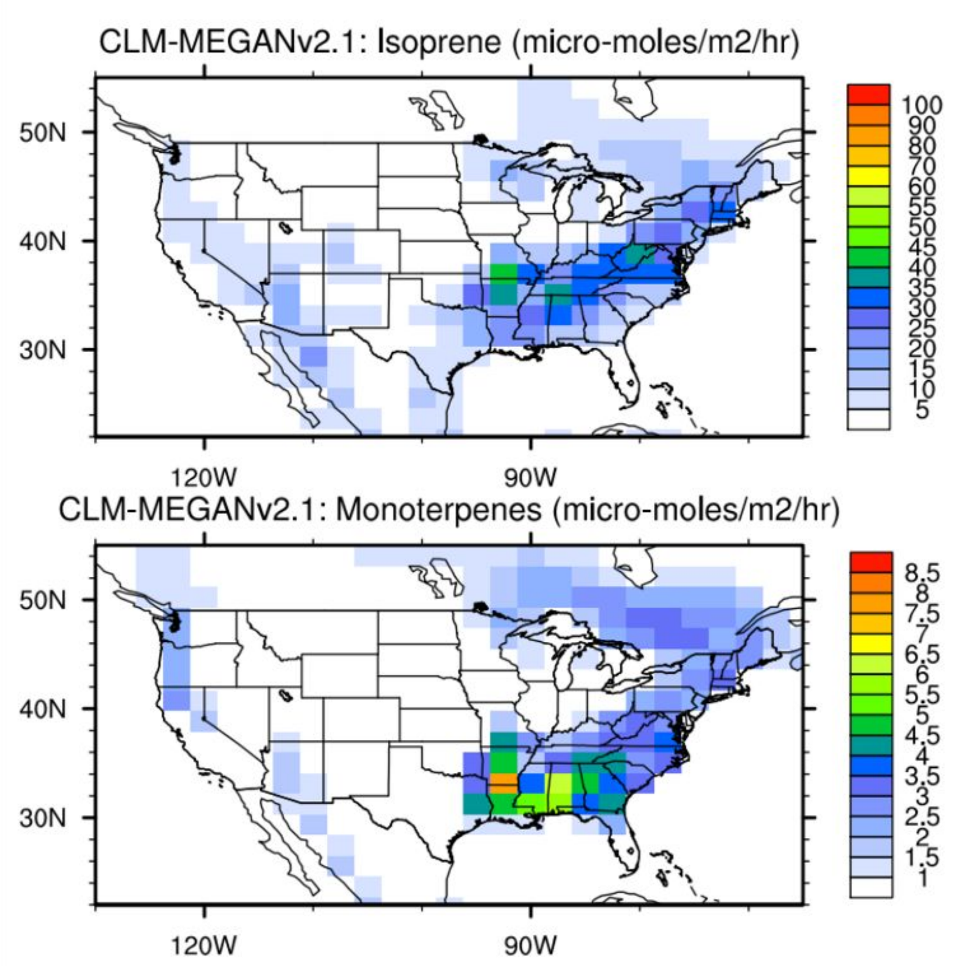
$$F_i = \gamma_i \ \sum \varepsilon_{i,j} \, \chi_j$$

where

γ_i : emission activity factor, depends on **leaf area index (LAI)**, **meteorology** (T, solar radiation), **leaf age**, with separate light-dependent and light-independent factors

$\varepsilon_{i,j}$: emission factor at standard conditions for vegetation type (PFT) j

χ_j : fractional area of **PFT** j


CLM-MEGANv2.1: Isoprene (micro-moles/m2/hr)

CLM-MEGANv2.1: Monoterpenes (micro-moles/m2/hr)

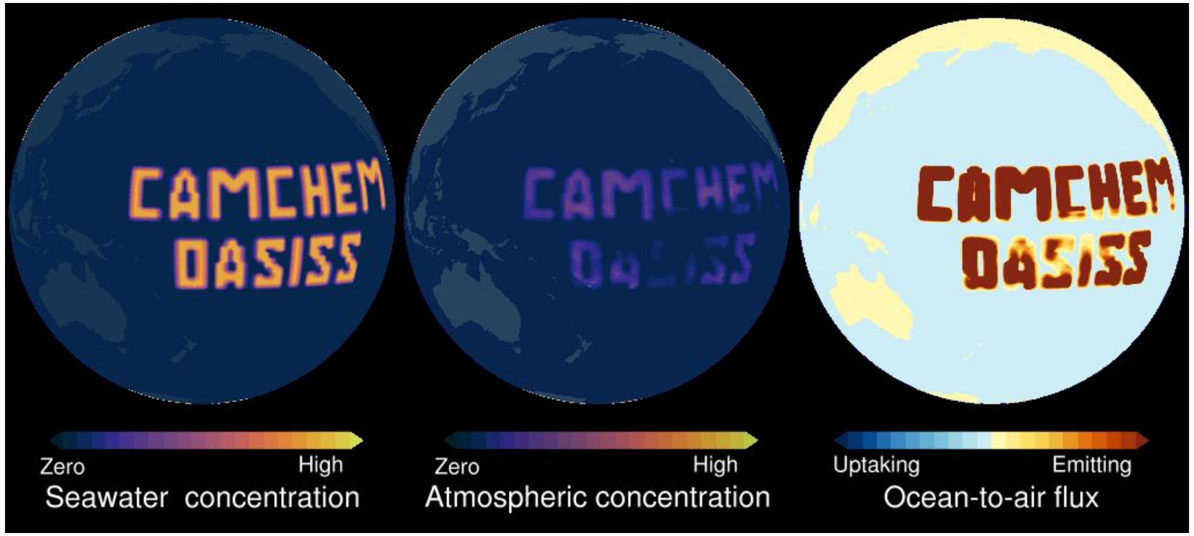
Guenther et al., GMD, 2012; https://gmd.copernicus.org/articles/5/1471/2012/

Interactive emissions: Ocean DMS

DMS emissions from ocean are calculated online based on the Online Air-Sea Interface for Soluble Species (OASISS) module:

https://wiki.ucar.edu/pages/viewpage.action?pageId=358319521

Seawater concentrations are specified and the emissions flux is calculated each timestep based on the model winds, etc.



Wang, S.,et al. (2020). JGR-Atmos.
https://doi.org/10.1029/2020JD032553

Interactive emissions: Biomass Burning in Land Component (CLM) (experimental)

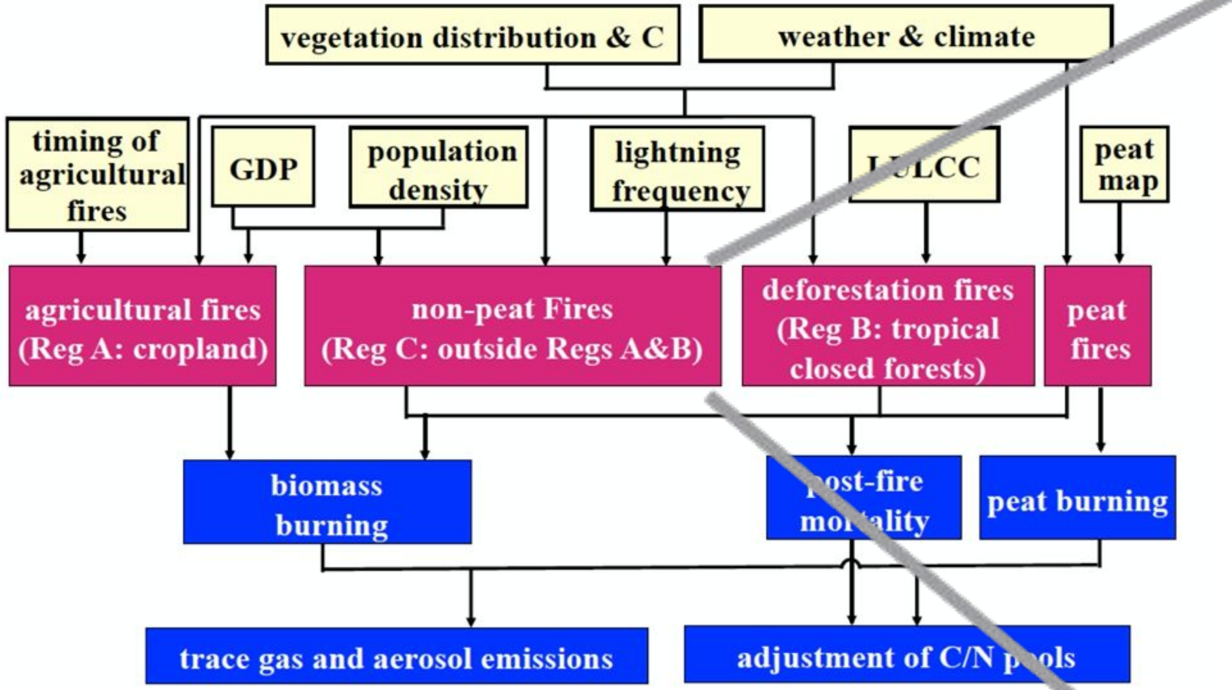


Fig. 2. Structure of new fire parameterization. Fire scheme described in Li et al. (2012a, b) is used in Region C with modifications by mainly adding the economic influence in the fire occurrence component and the socioeconomic influence in the fire spread component.

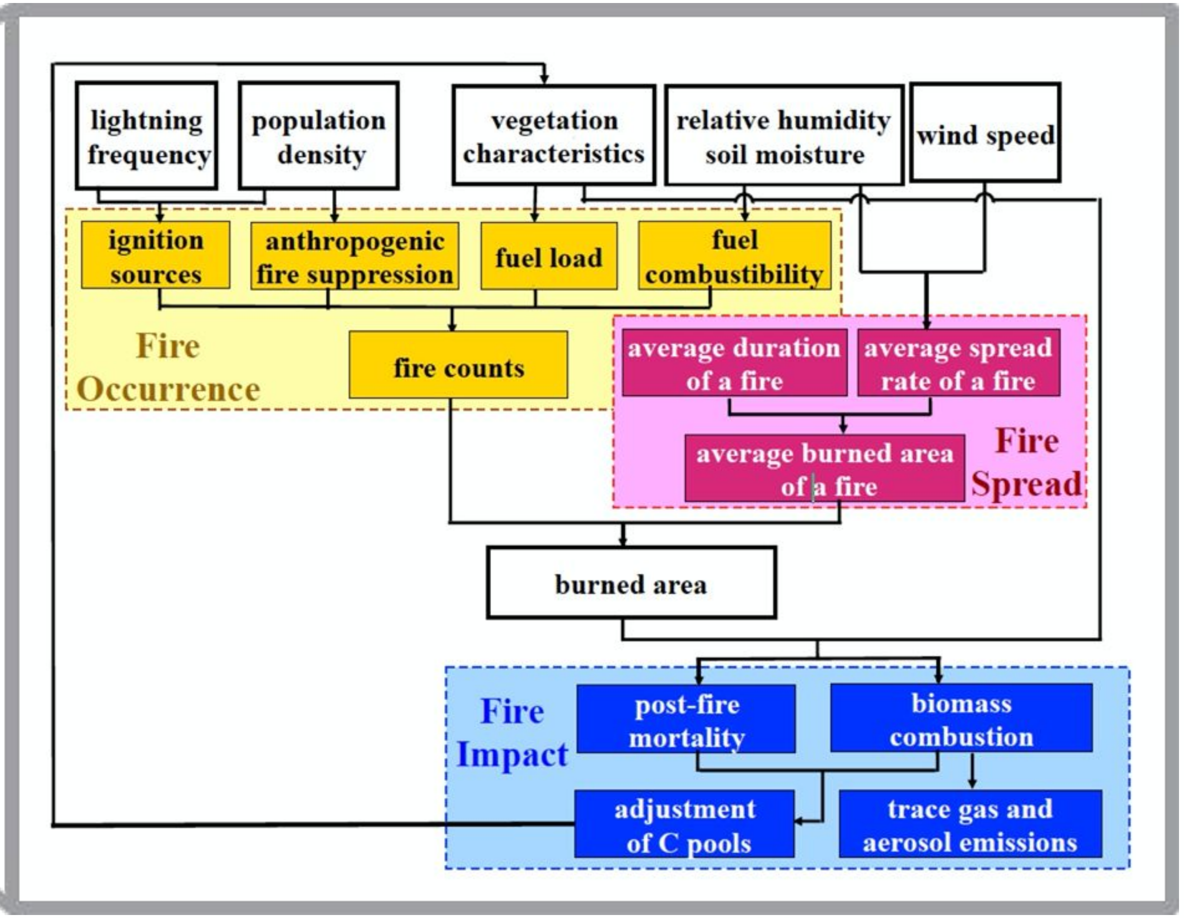


Fig. 1. Fire parameterization of Li et al. (2012a, b). It contains three components: fire occurrence, fire spread, and fire impact.

Li et al., 2012, 2013

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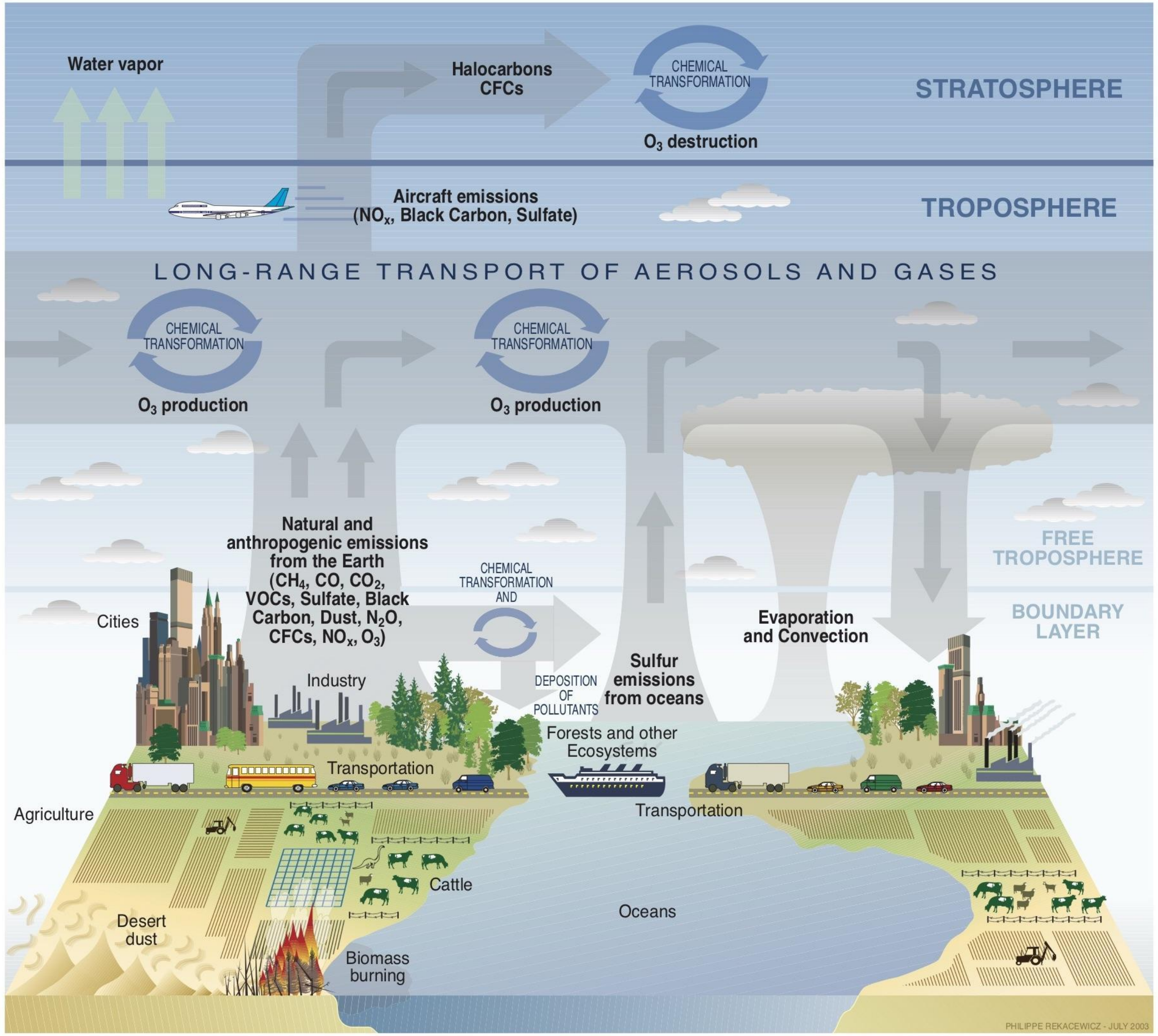

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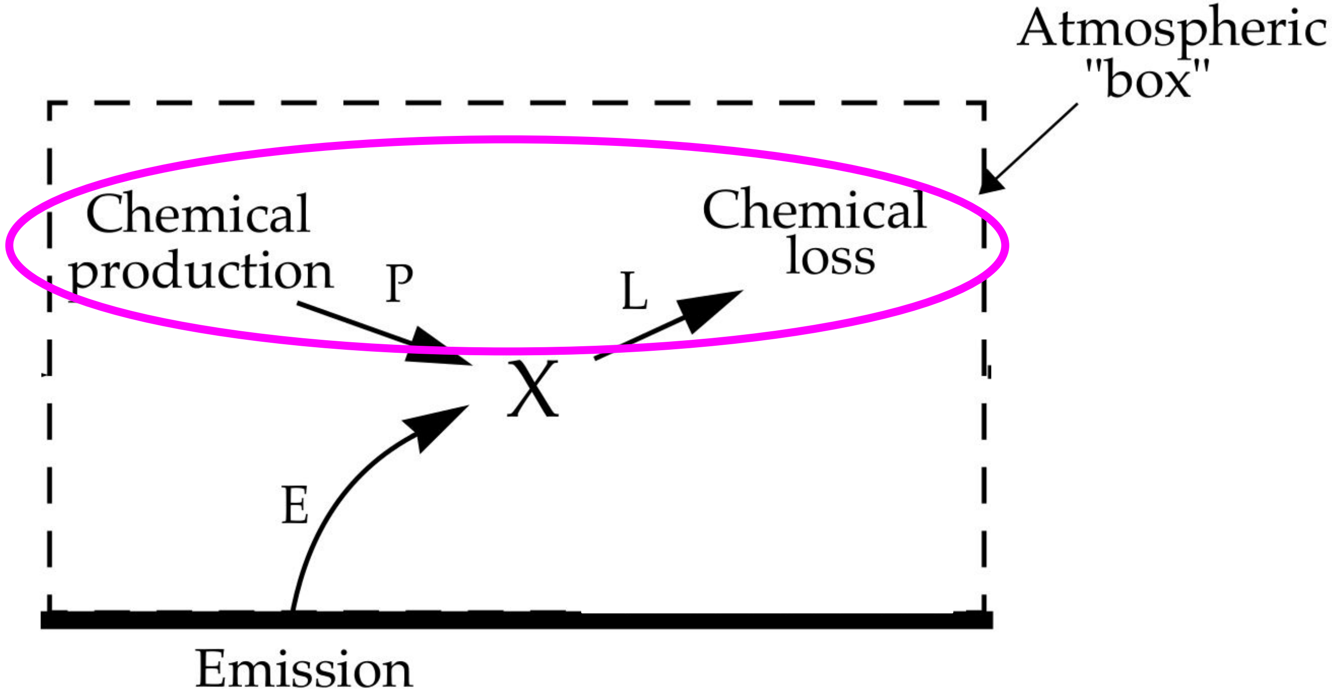
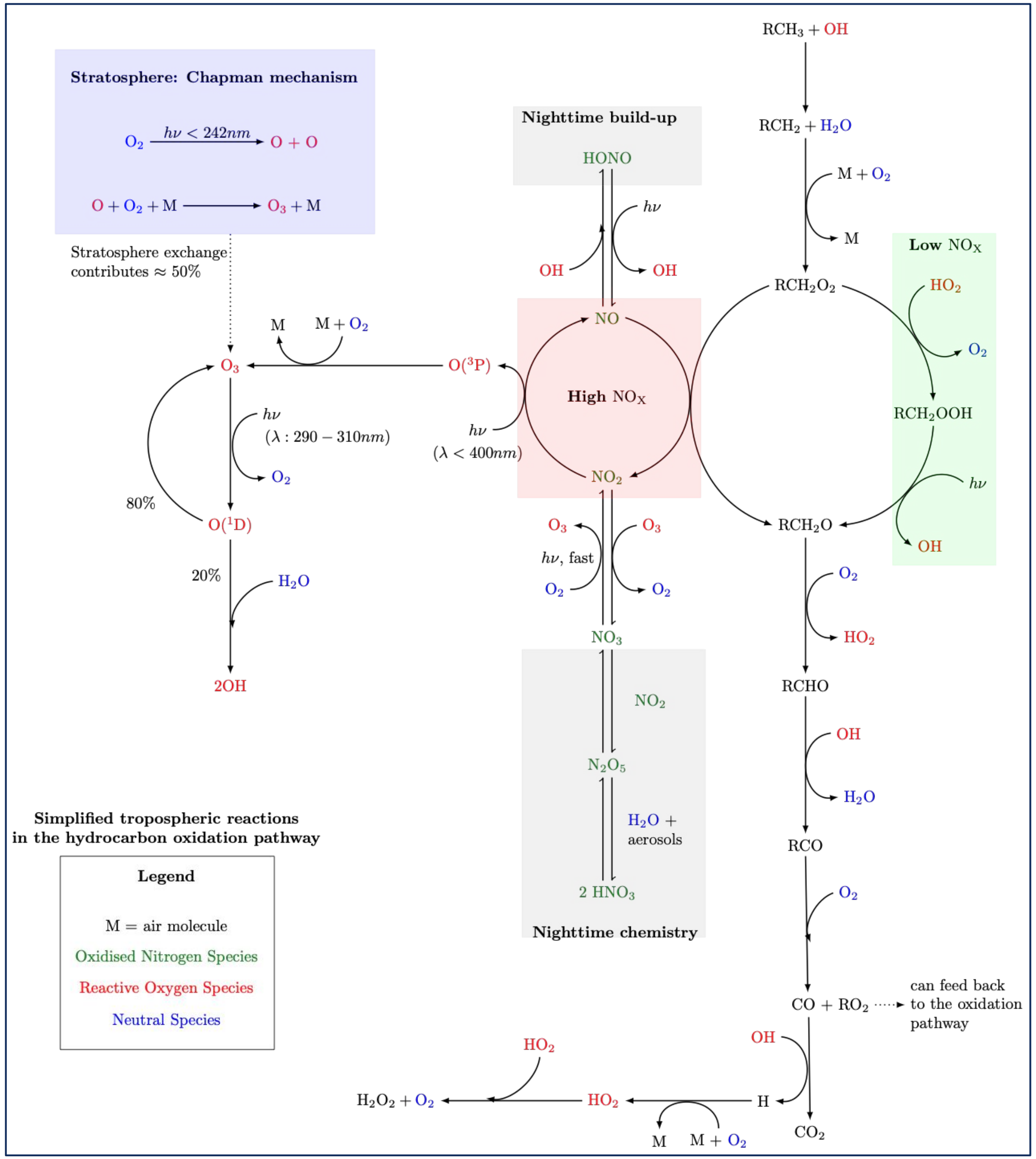
E_i Emissions
C_i Gas-phase-Chemistry
A_i Aerosol-processes
(Gas-aerosol exchange,
het chem.)



Introduction to Atmospheric Chemistry, Daniel J. Jacob https://acmg.seas.harvard.edu/education/introduction-atmospheric-chemistry

Atmospheric chemistry

- Time and light are important: photochemical reactions change and remove emissions
  - OH radical is the main atmospheric “detergent”
  - Can sometimes make more harmful species e.g. ozone (O_3)
- Emitted species undergo transport, deposition and chemical loss/transformation

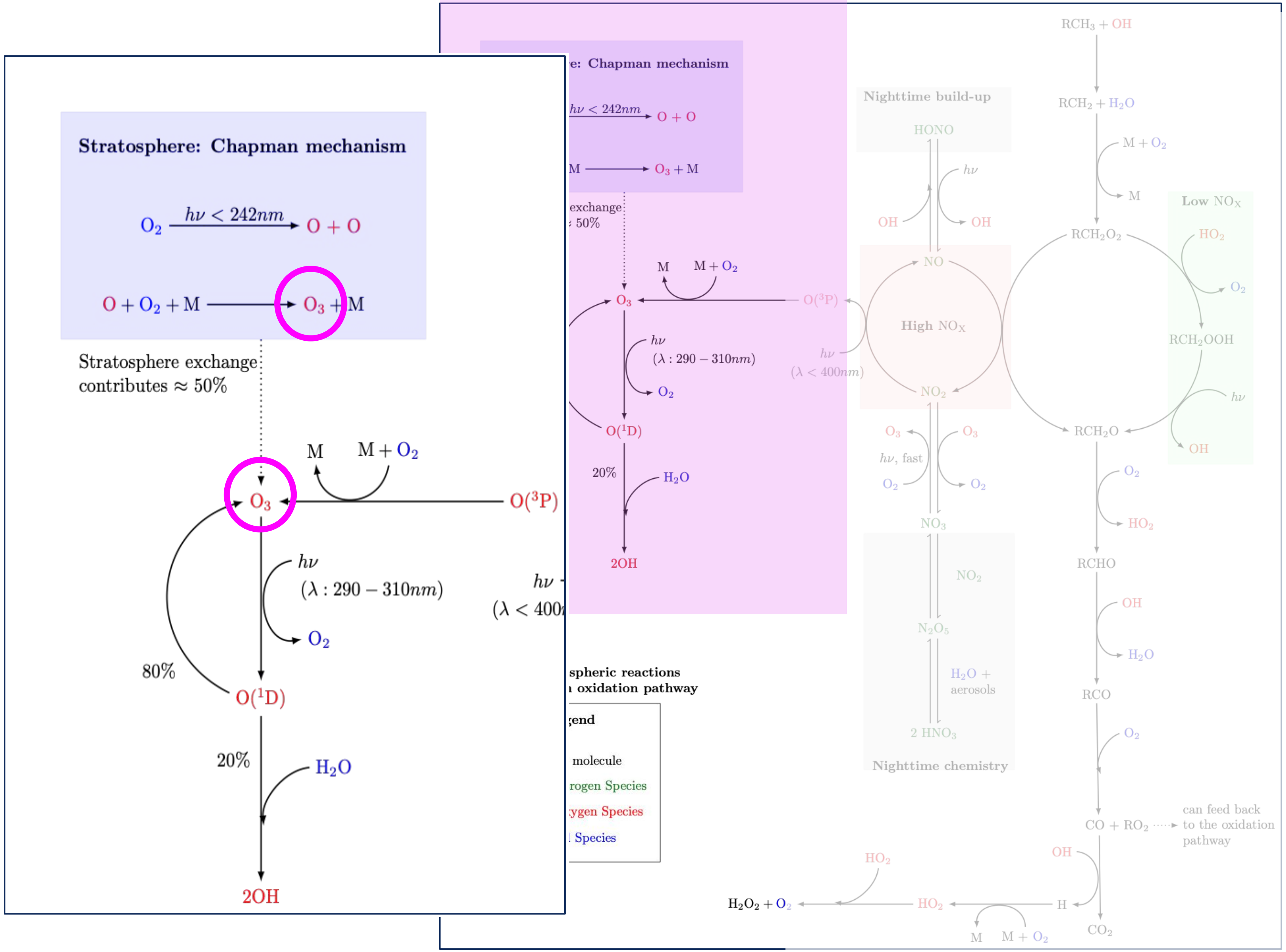


Simplified Hydrocarbon Oxidation Pathway

https://doi.org/10.6084/m9.figshare.7076282.v1

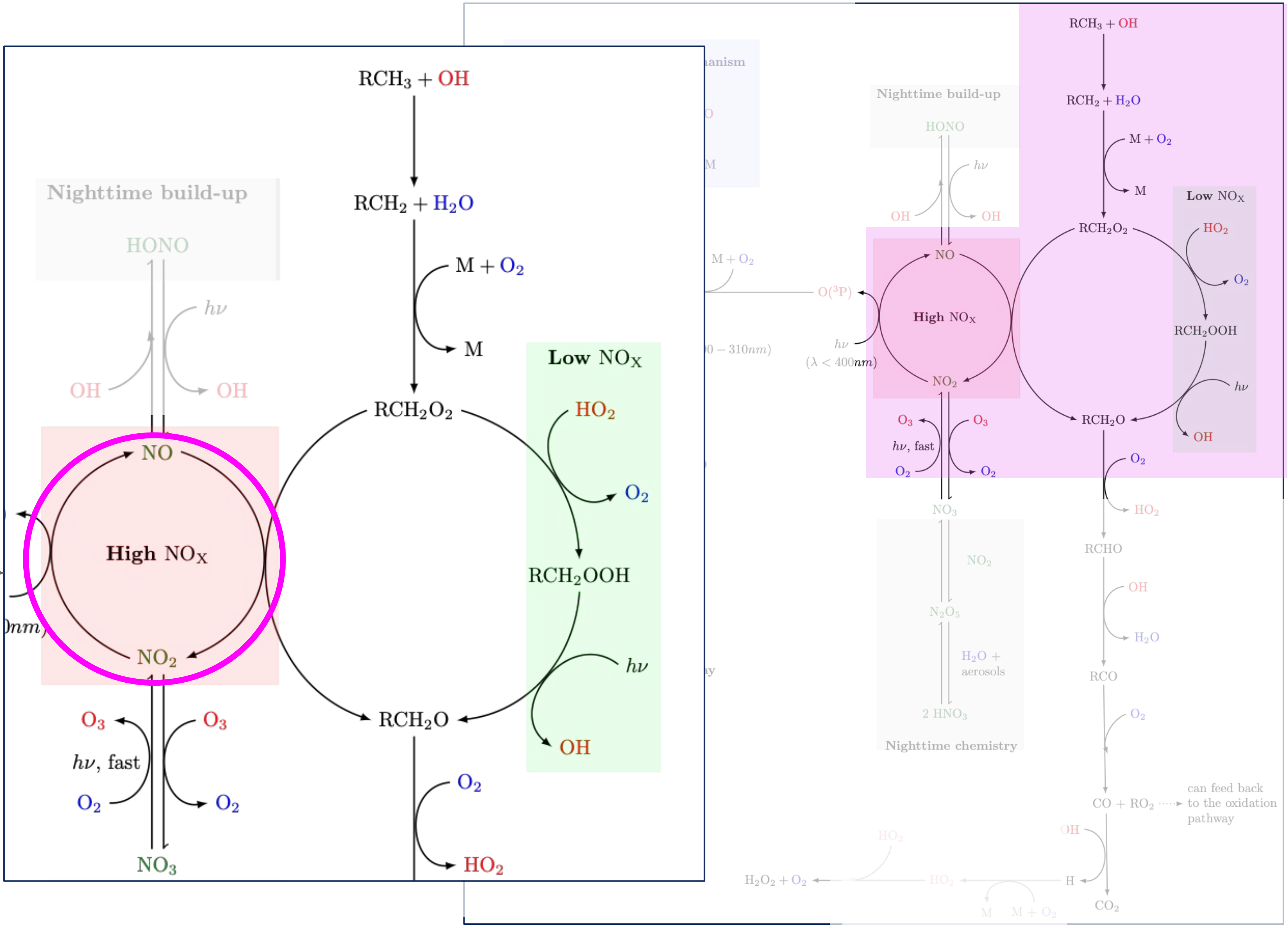
Stratospheric ozone

Tropospheric ozone



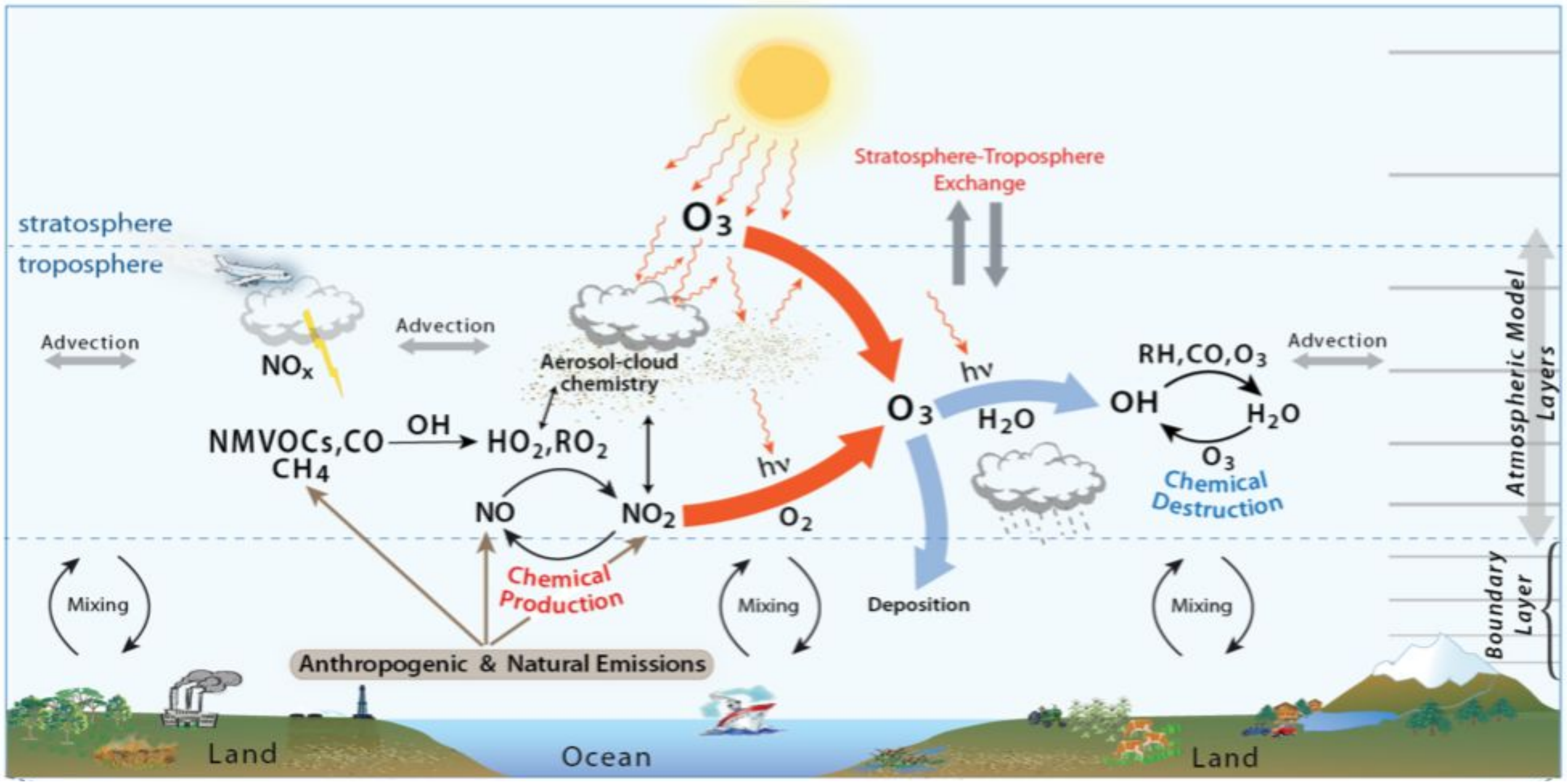
Shows high and low NOx pathways

NO_X cycle



Tropospheric Chemistry

Photochemistry
Gas-phase chemistry
Heterogeneous chemistry
Aqueous phase chemistry
Gas-to-aerosol exchange



Young et al., 2017

Atmospheric Chemistry

Explicit/Comprehensive Chemistry examples:

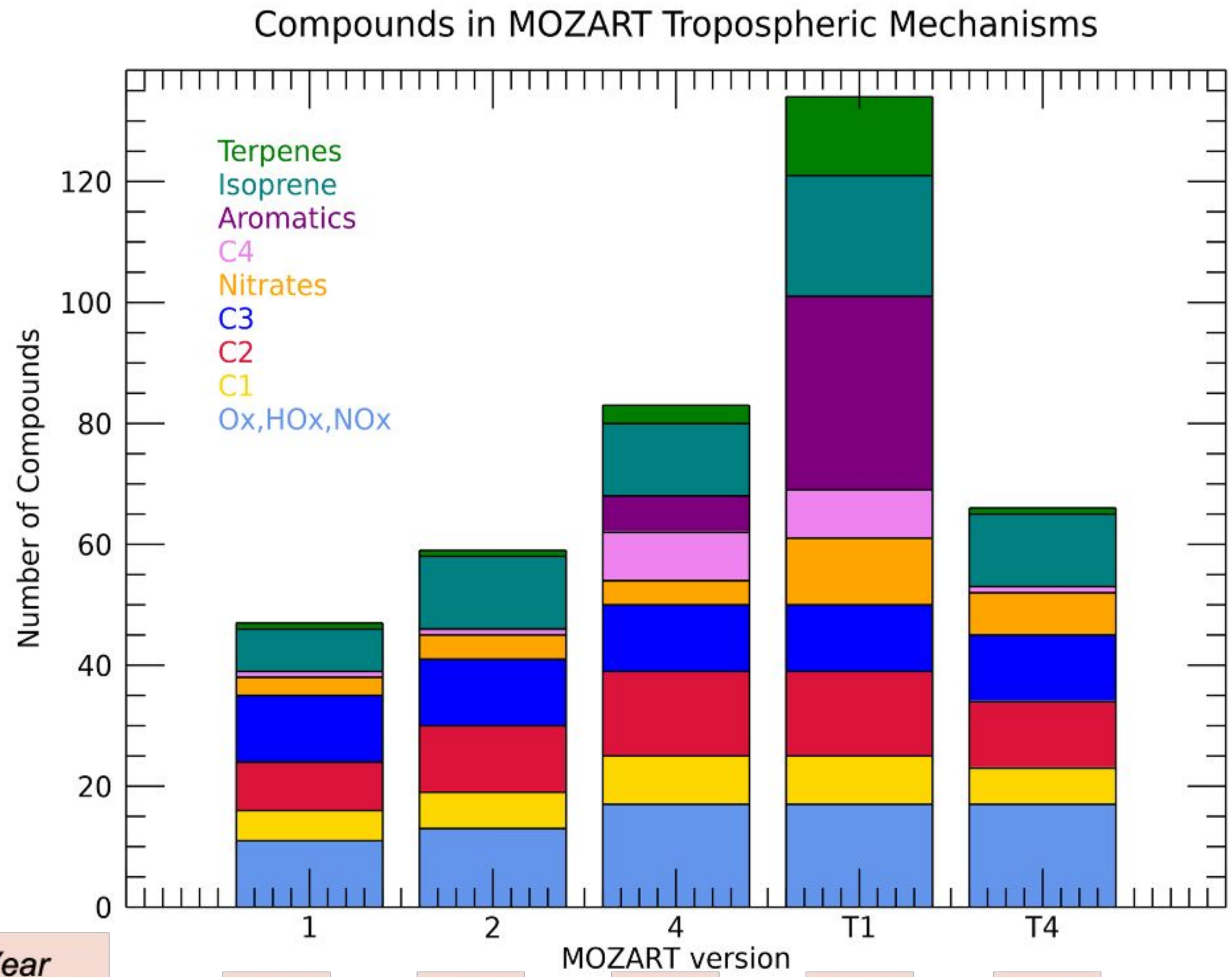
- Master Chemical Mechanism (MCM): 17224 reactions comprising 5832 different species (https://mcm.york.ac.uk/MCM/)
- Generator of Explicit Chemistry and Kinetics for Organics in the Atmosphere (GECKO-A): uses structure – reactivity relationships to create detailed chemical schemes for different environments (urban, rural etc.) (https://www2.acom.ucar.edu/modeling/gecko)

Global Earth System Models:

- known chemistry needs simplifying/condensing → e.g. lumping higher alkanes
- balance between computational efficiency and chemical accuracy
- historically: start with simplified mechanism and build complexity
- choose the chemistry to answer the scientific question

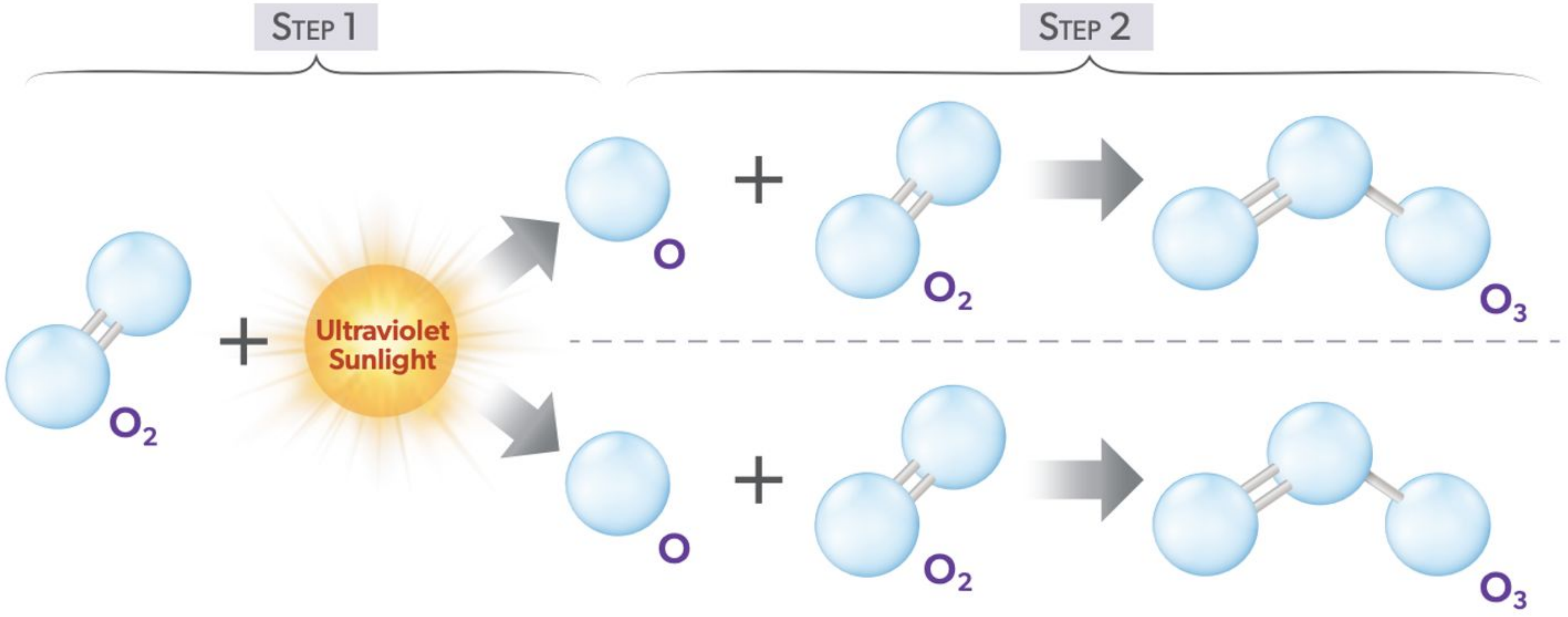
MOZART Family of Chemical Mechanisms

- Increasing complexity as computing power increased
- The MOZART-T4 mechanism is comparable to MOZART-2 (Horowitz et al., 2003)
- Similar mechanism used in GFDL AM4 (Horowitz et al., 2019)
- MOZART-T4 not optimal for air quality studies, but should appropriately simulate oxidants and aerosols for chemistry-climate studies and for creating specified oxidants for CAM



Year developed 1998 2003 2010 2020 2024

Stratospheric Ozone Production



OVERALL REACTION : $3\,O_2 \xrightarrow{\text{SUNLIGHT}} 2\,O_3$

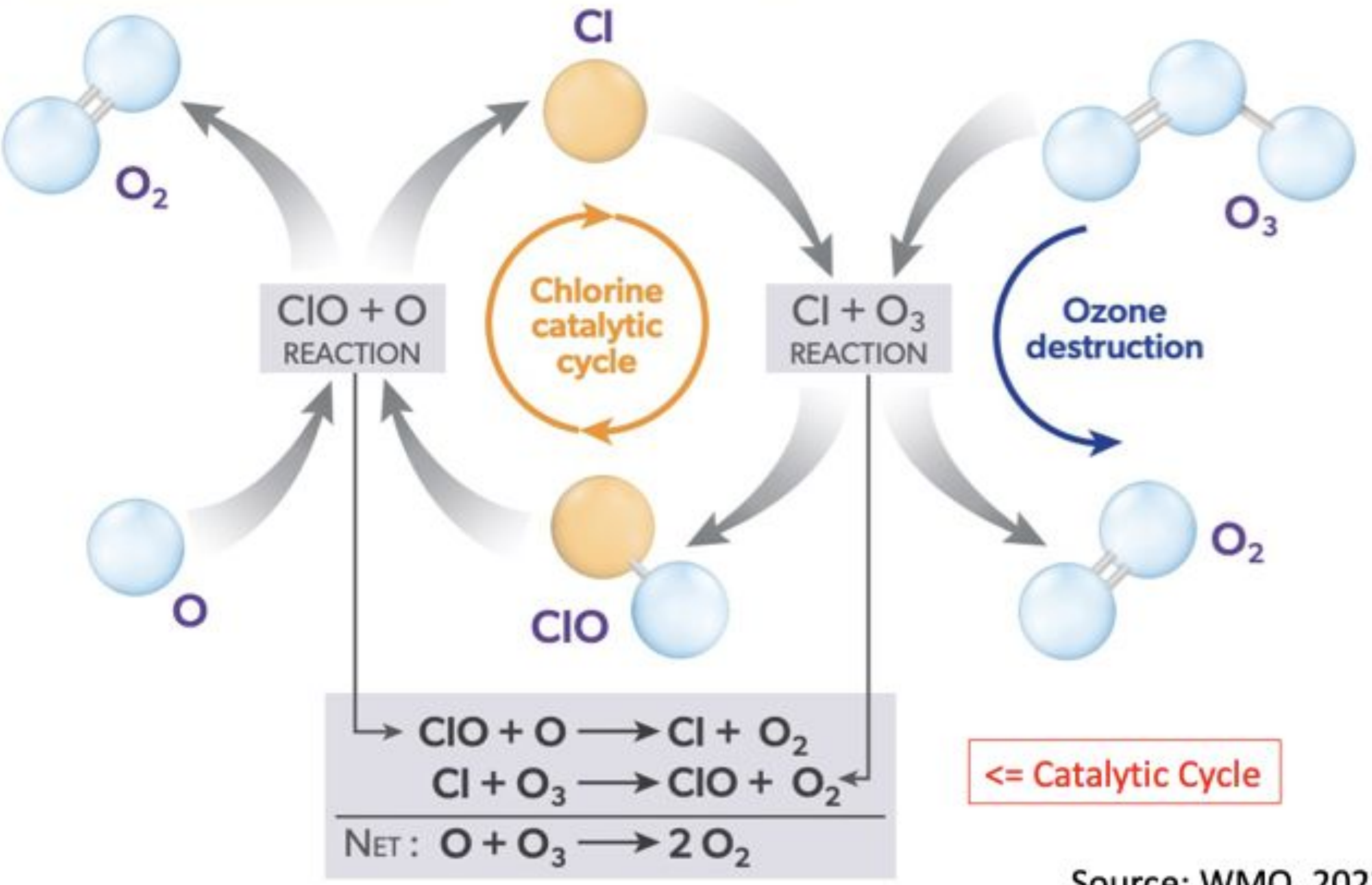
Figure Q1-3. Stratospheric ozone production. Ozone is produced naturally in the stratosphere by a two-step reaction process. In the first step, solar ultraviolet radiation (sunlight) breaks apart an oxygen molecule to form two separate oxygen atoms. In the second step, each oxygen atom collides with another oxygen molecule and forms an ozone molecule in a binding reaction. In the overall process, three oxygen molecules plus sunlight react to form two ozone molecules.

Industrial production of CFCs, HCFCs (Solvents, Refrigeration, Foam Blowing) have a long atmospheric lifetime (10-100 years)

Destruction in Stratosphere

Release Chlorine Atom

Ozone Destruction Cycle 1 : Upper Stratosphere



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

NET : $O + O_3 \longrightarrow 2\,O_2$

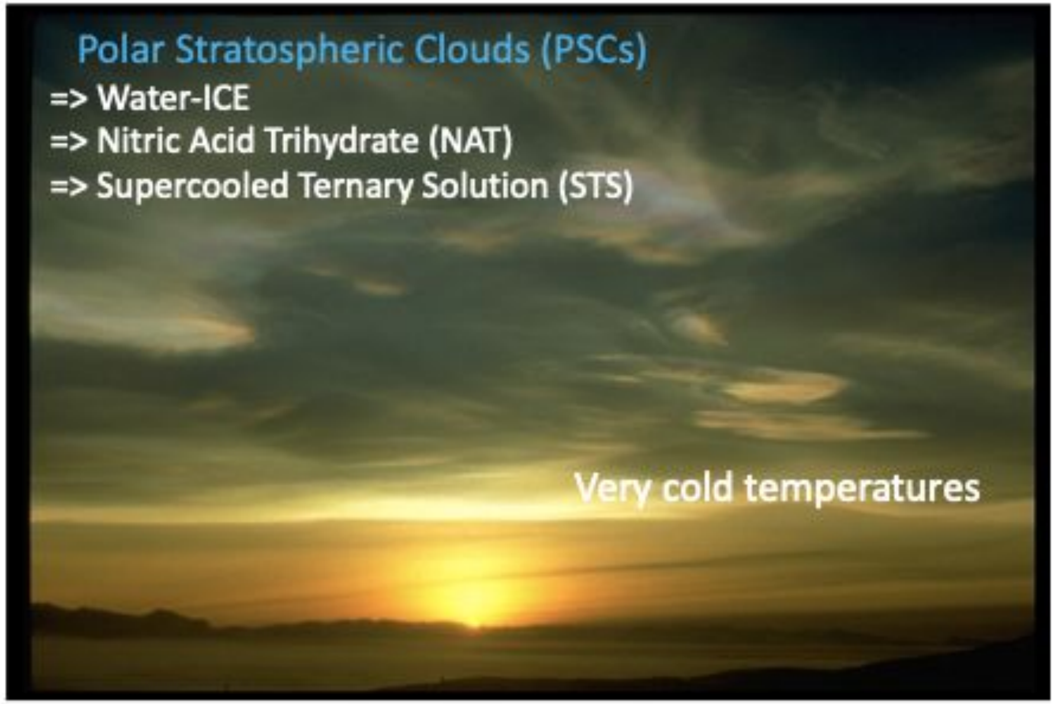
<= Catalytic Cycle

Source: WMO, 2022

In the polar regions chlorine species are primarily in reservoir form (e.g., HCl and $ClONO_2$). [Not very reactive with ozone]

Converted to active form on the surface of polar stratospheric clouds (see below).

$HCl + ClONO_2 \Rightarrow Cl_2 + HNO_3 \Rightarrow 2ClO$



Polar Stratospheric Clouds (PSCs)

=> Water-ICE

=> Nitric Acid Trihydrate (NAT)

=> Supercooled Ternary Solution (STS)

Very cold temperatures

Ozone Destruction Cycles 2 and 3 : Polar Regions

CYCLE 2 :

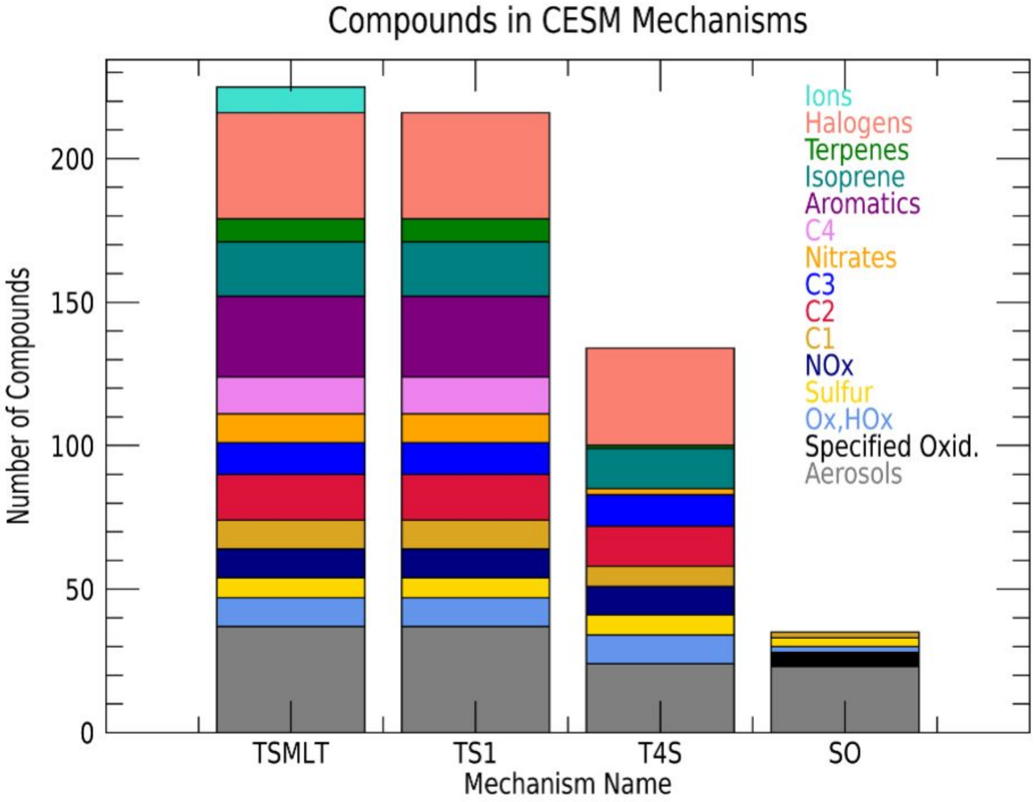
$$\begin{aligned} ClO + ClO &\rightarrow (ClO)_2 \\ (ClO)_2 + \textit{sunlight} &\rightarrow ClOO + Cl \\ ClOO &\rightarrow Cl + O_2 \\ 2(Cl + O_3 &\rightarrow ClO + O_2) \\ \hline \text{NET: } 2O_3 &\rightarrow 3O_2 \end{aligned}$$

Another example of a Catalytic Cycle

Source: WMO, 2022

Atmospheric chemistry mechanisms in CESM

Chemistry mechanism descriptions:
https://www2.acom.ucar.edu/gcm/mozart



Name	Description	# tracers	#rxns
T1MA (TSMLT)	T1 with stratosphere, mesosphere, lower thermosphere chemistry	234	583
T1S	T1 with comprehensive stratospheric chemistry and full sulfur chemistry	231	528
T4S	T4 with comprehensive stratospheric chemistry, no odd F, C>3 hydrocarbons simplified	141	*
SO	Specified Oxidants, with GHGs	31	12

T1S= default
“full-chemistry”
Troposphere and Stratosphere

CAM6 vs CAM-chem

Same atmosphere, physics, resolution

Different chemistry and aerosols -> emissions and coupling

- **CAM6:** Aerosols are calculated, using simple chemistry (“fixed” oxidants) (prescribed: N_2, O_2, H_2O, O_3, OH, NO_3, HO_2; chemically active: H_2O_2, H_2SO_4, SO_2, DMS, SOAG)

  Limited interactions between Chemistry and Climate

  -> prescribed fields are derived using chemistry-climate simulations

  - Prescribed ozone is used for radiative calculations
  - Prescribed oxidants is used for aerosol formation
  - Prescribed methane oxidation rates
  - Prescribed stratospheric aerosols
  - Prescribed nitrogen deposition
  - Simplified secondary organic aerosol description

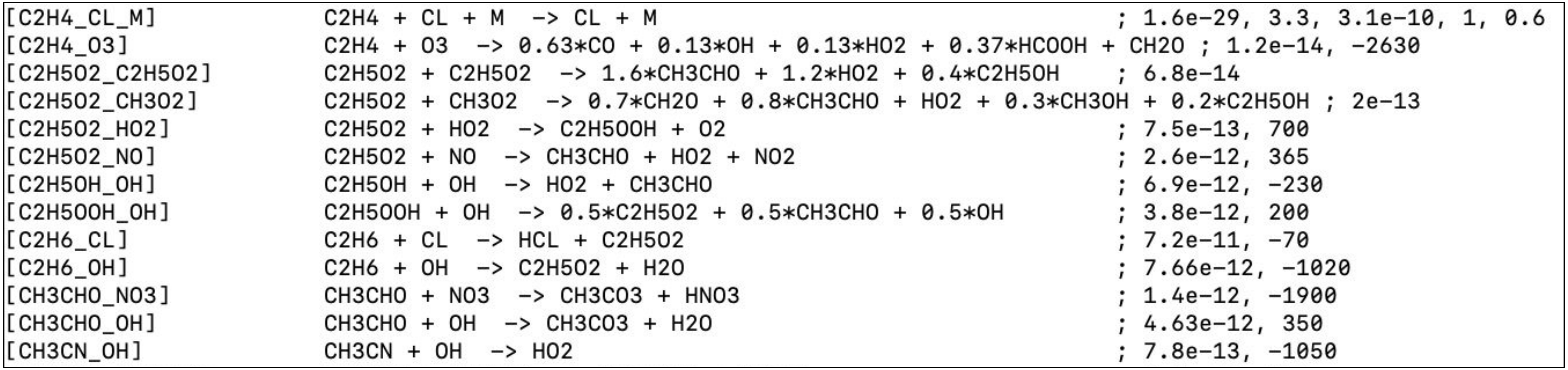
Example of chemistry mechanism code

text file: chem_mech.in

read into the chemistry preprocessor

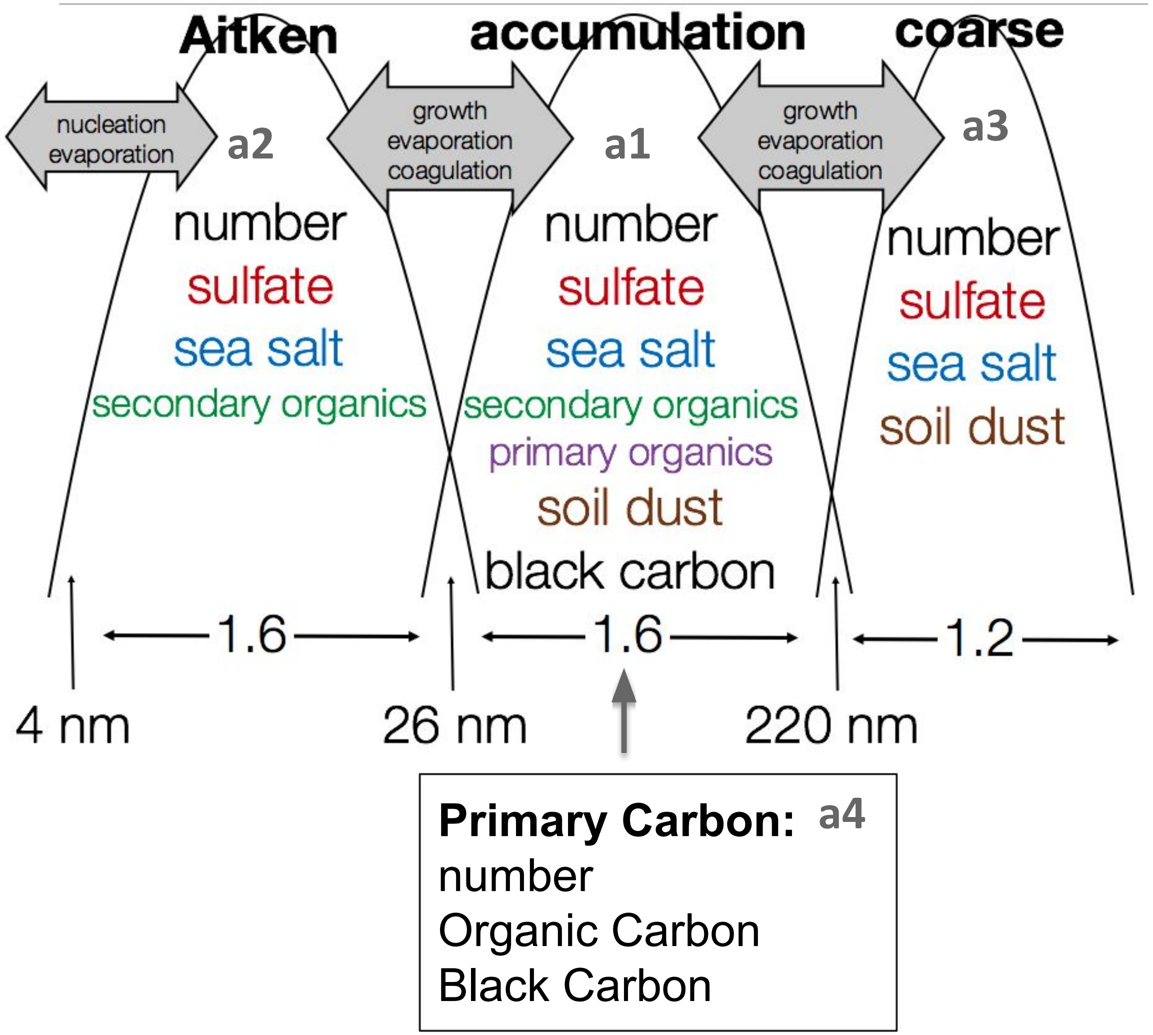
Reaction rates

Different reaction formats depending on whether temperature and air density are important.



```
[C2H4_CL_M]            C2H4 + CL + M  -> CL + M                                   ; 1.6e-29, 3.3, 3.1e-10, 1, 0.6
[C2H4_O3]              C2H4 + O3  -> 0.63*CO + 0.13*OH + 0.13*HO2 + 0.37*HCOOH + CH2O ; 1.2e-14, -2630
[C2H5O2_C2H5O2]        C2H5O2 + C2H5O2  -> 1.6*CH3CHO + 1.2*HO2 + 0.4*C2H5OH     ; 6.8e-14
[C2H5O2_CH3O2]         C2H5O2 + CH3O2  -> 0.7*CH2O + 0.8*CH3CHO + HO2 + 0.3*CH3OH + 0.2*C2H5OH ; 2e-13
[C2H5O2_HO2]           C2H5O2 + HO2  -> C2H5OOH + O2                              ; 7.5e-13, 700
[C2H5O2_NO]            C2H5O2 + NO  -> CH3CHO + HO2 + NO2                         ; 2.6e-12, 365
[C2H5OH_OH]            C2H5OH + OH  -> HO2 + CH3CHO                               ; 6.9e-12, -230
[C2H5OOH_OH]           C2H5OOH + OH  -> 0.5*C2H5O2 + 0.5*CH3CHO + 0.5*OH          ; 3.8e-12, 200
[C2H6_CL]              C2H6 + CL  -> HCL + C2H5O2                                 ; 7.2e-11, -70
[C2H6_OH]              C2H6 + OH  -> C2H5O2 + H2O                                 ; 7.66e-12, -1020
[CH3CHO_NO3]           CH3CHO + NO3  -> CH3CO3 + HNO3                             ; 1.4e-12, -1900
[CH3CHO_OH]            CH3CHO + OH  -> CH3CO3 + H2O                               ; 4.63e-12, 350
[CH3CN_OH]             CH3CN + OH  -> HO2                                         ; 7.8e-13, -1050
```

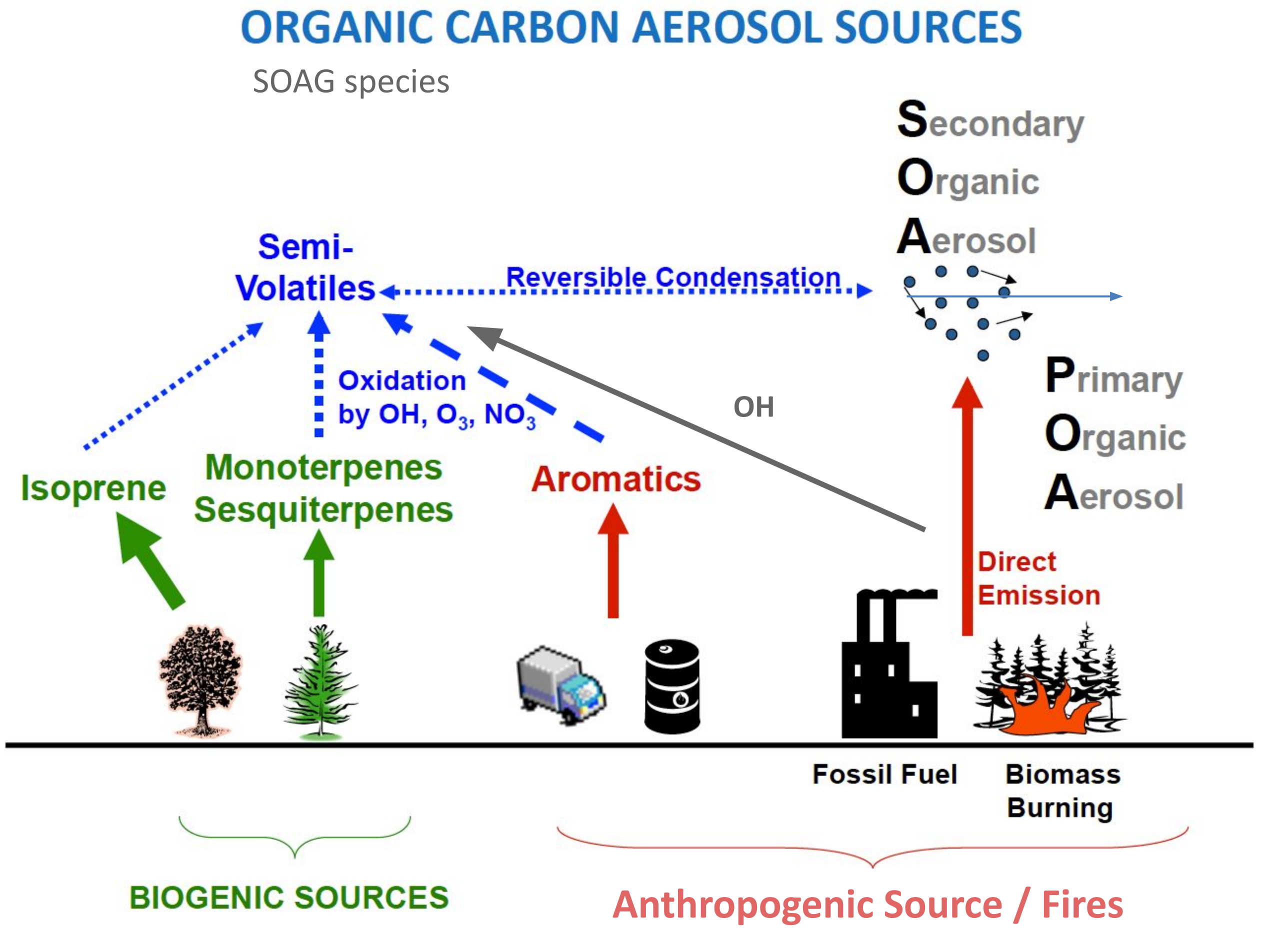
Default Modal Aerosol Model (MAM4)



Representation of
- Sulfates,
- Black Carbon
- Organic Carbon, Organic Matter (OC, SOA),
- Mineral Dust and Sea-Salt

Liu et al., 2016 *Courtesy Mike Mills*

Secondary Organic Aerosol Description



Modified from C. Heald, MIT Cambridge

Simplified Chemistry (CAM6):

- SOAG (oxygenated VOCs) derived from fixed mass yields
- no interactions with land

Comprehensive Chemistry:

- SOAG formation derived from VOCs using Volatility Bin Set (VBS)
- 5 volatility bins
- Interactive with land emissions

-> a more physical approach

Atmospheric Chemistry

- Motivation
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    - Chemical mechanism
    - Aerosol model and cloud interactions
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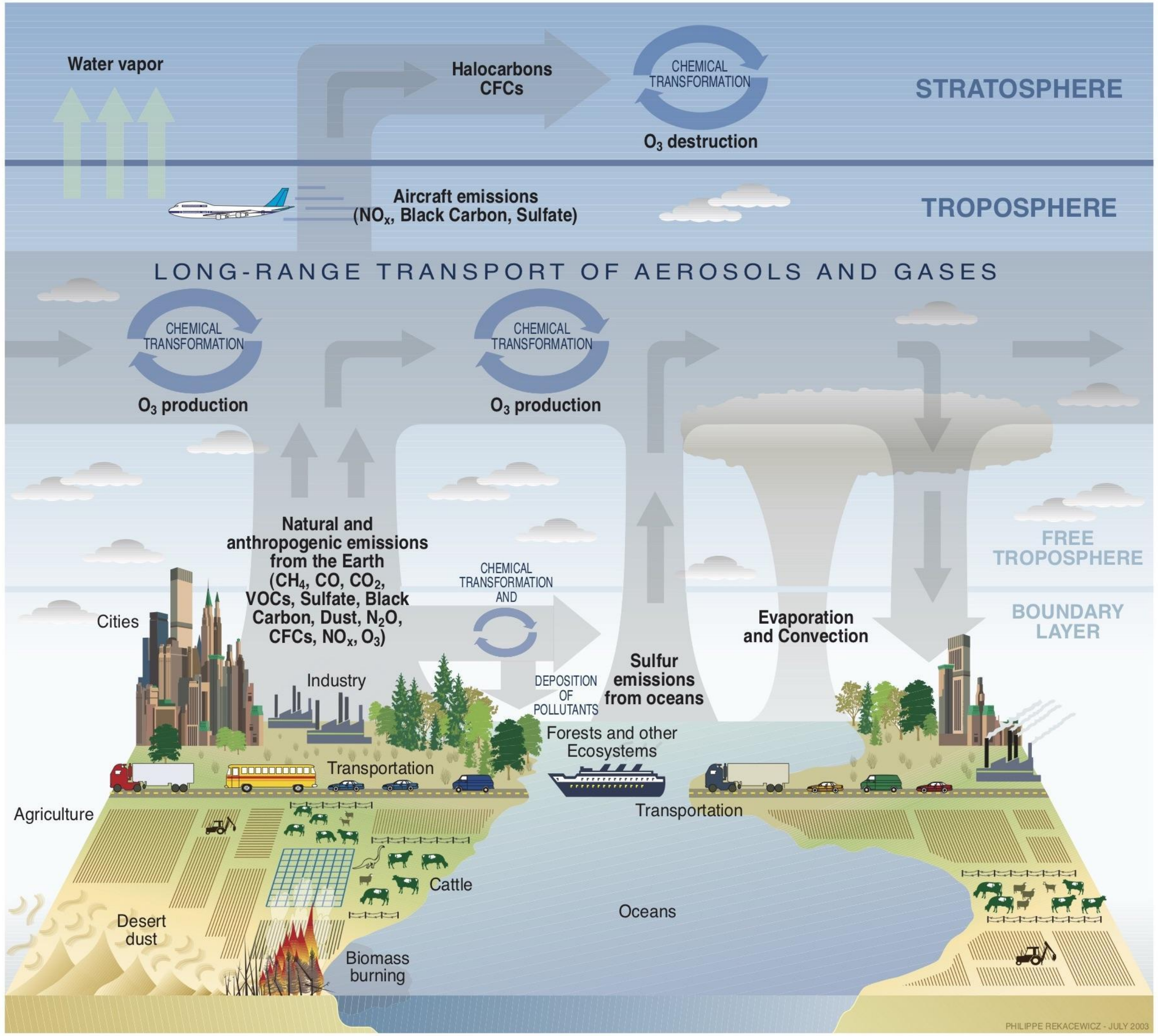
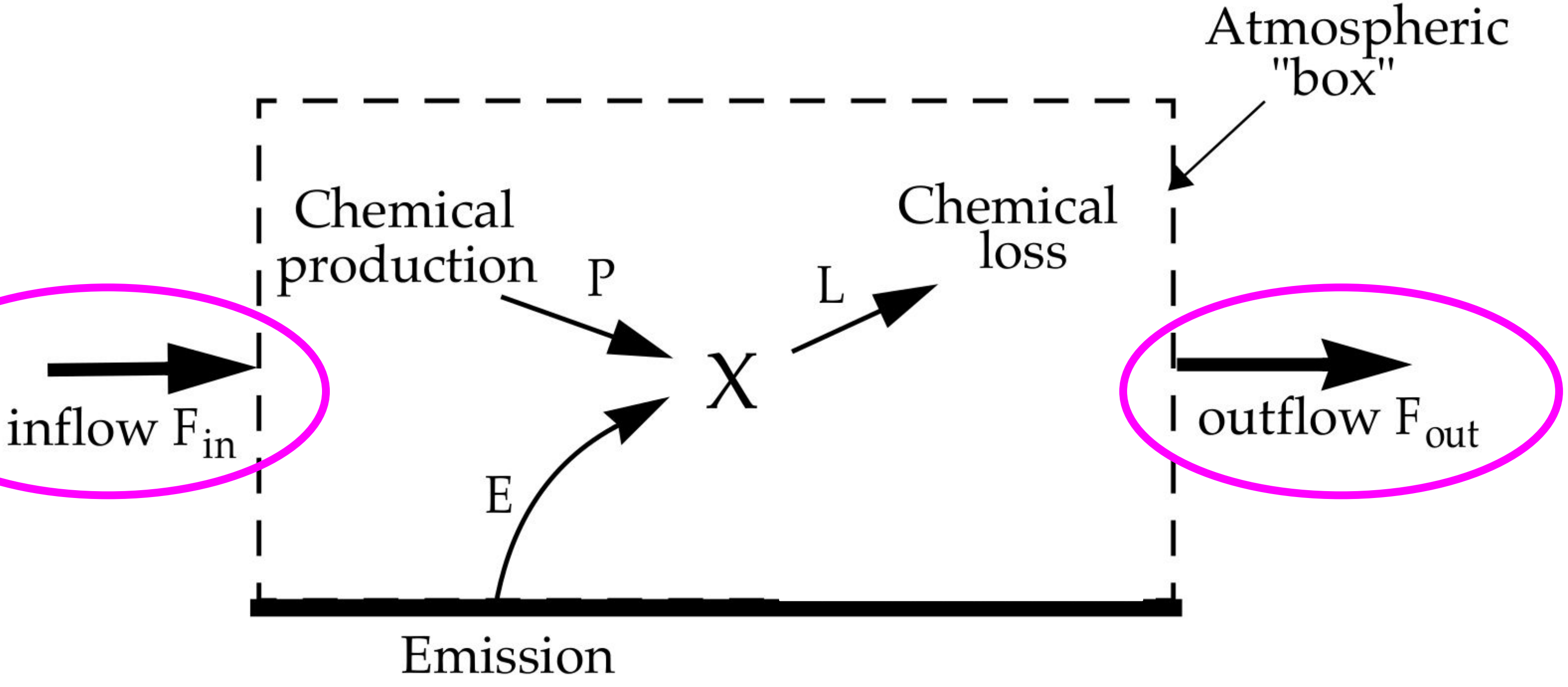


For each chemical constituent (χ), the following must be solved

$$\frac{\partial \chi(i)}{\partial t} = \mathrm{Sources(i)} - \mathrm{Sinks(i)} = \mathrm{E_i + C_i + A_i + T_i - W_i - D_i}$$

- ***E**$_i$* Emissions
- ***C**$_i$* Gas-phase-Chemistry
- ***A**$_i$* Aerosol-processes (Gas-aerosol exchange, het chem.)
- ***T**$_i$* Advection + Diffusion



Free running versus nudged (T, U, V)

Introduction to Atmospheric Chemistry, Daniel J. Jacob https://acmg.seas.harvard.edu/education/introduction-atmospheric-chemistry

Dynamical core overview

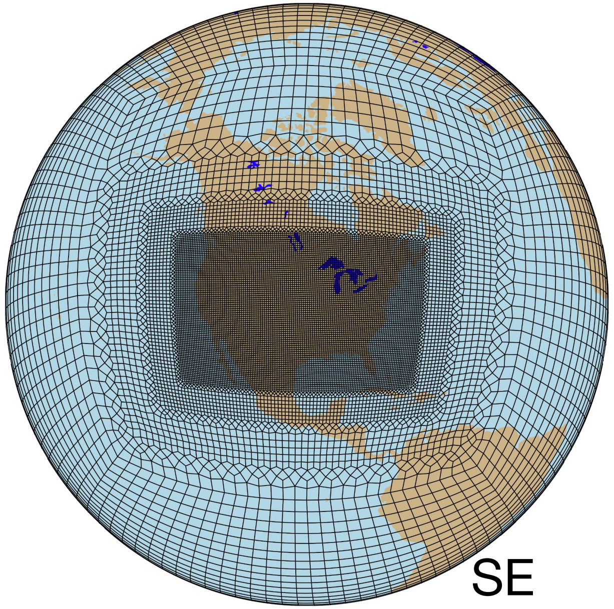
FV: Finite Volume (FV) “regular grid”

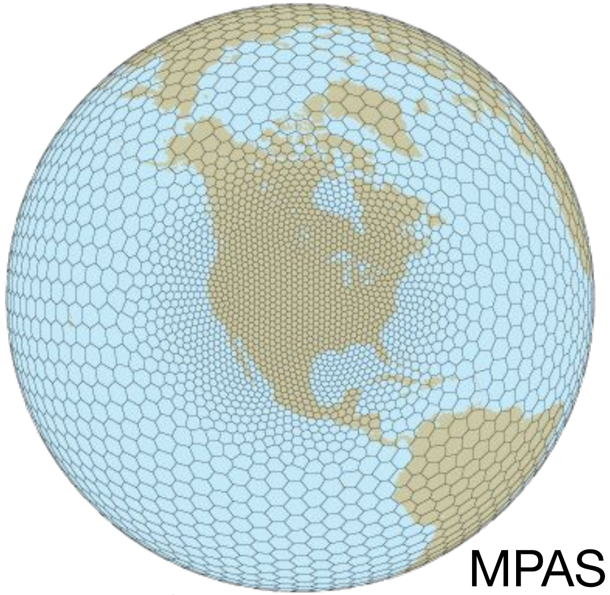
FV3: a non-hydrostatic cubed-sphere version of FV

SE - CSLAM (pg3): Spectral Element dynamical core on a cubed sphere, Conservative Semi-Lagrangian Multi-tracer dynamical core with finite-volume transport (CSLAM). No current regional refined capability.

SE (RR): Spectral Element dynamical core with regional refinement options.

MPAS: Model for Prediction Across Scales, cloud resolving, a global version of Weather Research and Forecasting, WRF, model discretized on a Voronoi grid. Regional refinement option, (experimental in CESM: need to compare with SE-RR).


SE


MPAS

https://www.cesm.ucar.edu/sites/default/files/2024-08/2024cesmtutoriallauritzen.pdf

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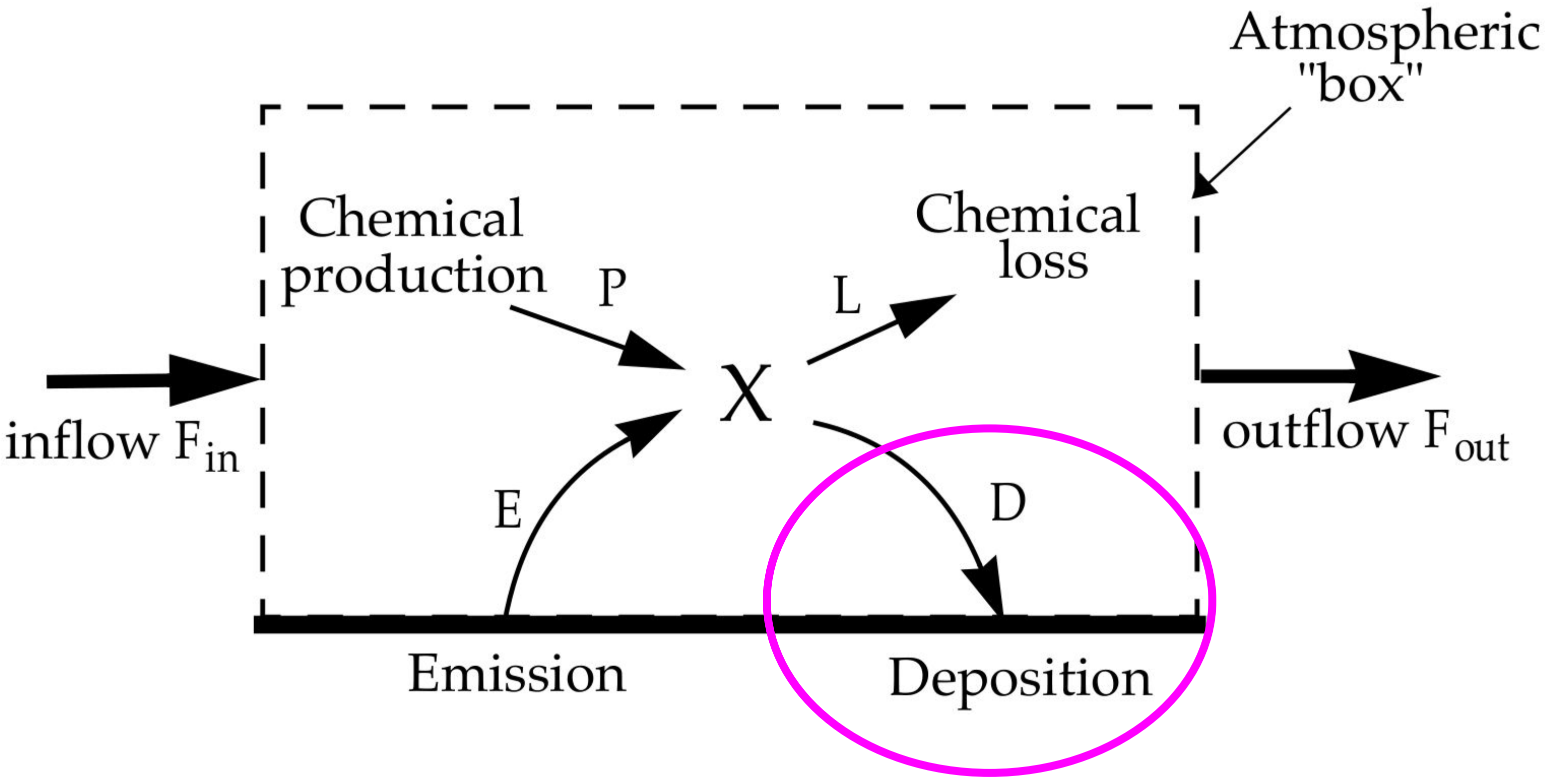
$$\frac{\partial \chi(i)}{\partial t} = \mathrm{Sources(i)} - \mathrm{Sinks(i)} = \mathrm{E_i} + \mathrm{C_i} + \mathrm{A_i} + \mathrm{T_i} - \mathrm{W_i} - \mathrm{D_i}$$

E_i	Emissions
C_i	Gas-phase-Chemistry
A_i	Aerosol-processes (Gas-aerosol exchange, het chem.)
T_i	Advection + Diffusion
W_i	Cloud-processes (wet deposition)
D_i	Dry deposition



Introduction to Atmospheric Chemistry, Daniel J. Jacob https://acmg.seas.harvard.edu/education/introduction-atmospheric-chemistry

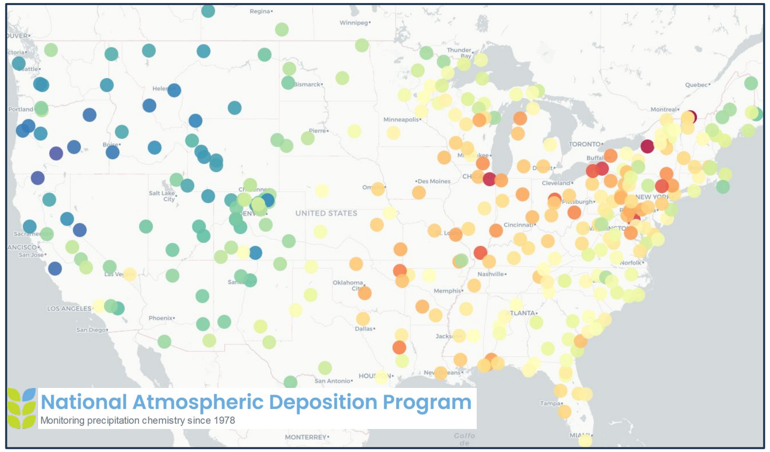
Wet Deposition

Large-scale and convective precipitation: uptake of chemical constituents in rain or ice

Considers in-cloud and below-cloud scavenging rates and solubility factors of aerosol and chemical species

A first-order loss process

$$\chi_{iscav} = \chi_i \times F \times (1 - \exp(-\lambda \Delta t))$$



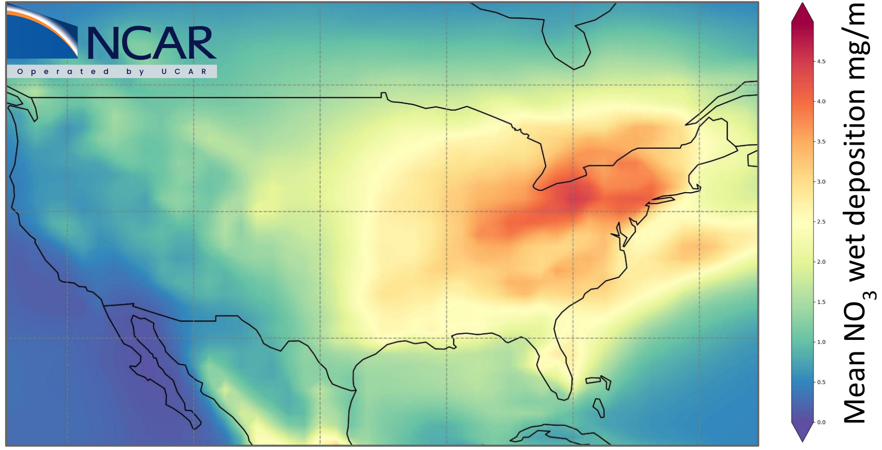
$\boldsymbol{X}_{iscav}$ scavenged species (kg)

$\boldsymbol{X}_i$ species

F fraction of the grid box from which tracer is being removed

$\boldsymbol{\lambda}$ is the loss rate

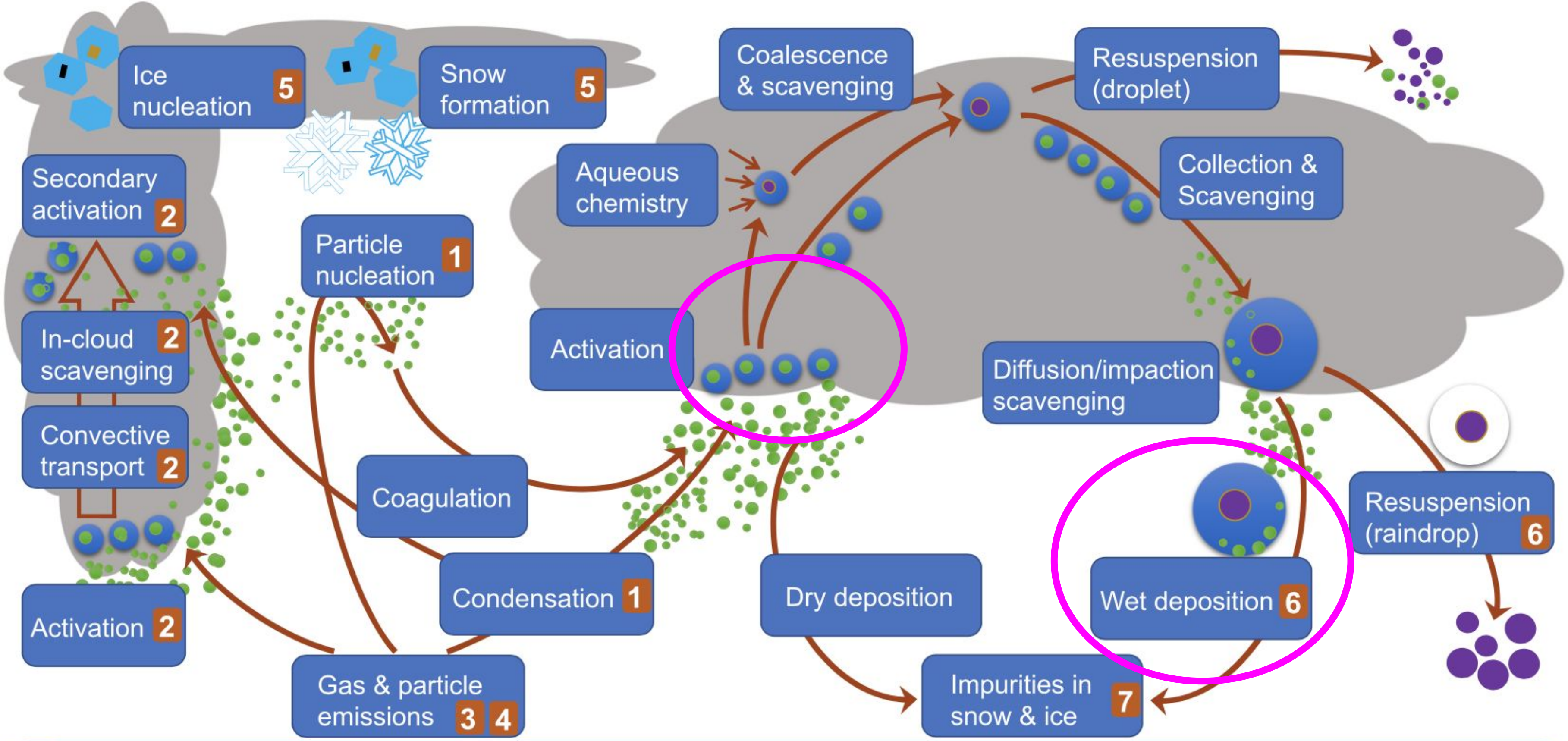
Deni Murray
ACOM ASP graduate visitor



References: (Barth et al., 2000, Neu and Prather 2012, Lamarque et al., 2012)

Aerosol – Cloud Interactions

Feedback into cloud condensation and precipitation



1 H_2SO_4 vapor production and loss (NewH2SO4)
2 Convective transport and wet removal (NewConvTran)
3 SOA precursor gas emission (NewSOA)
4 Representation of marine organic aerosol (NewMOA)
5 Ice nucleation and ice-to-snow conversion (NewIceNuc)
6 Resuspension of aerosol from evaporated raindrops (NewResusp)
7 Light-absorbing particles in snow and ice (NewInSnow)

E3SM: Wang et al., 2020 (JAMES)

Dry Deposition Velocity Calculation

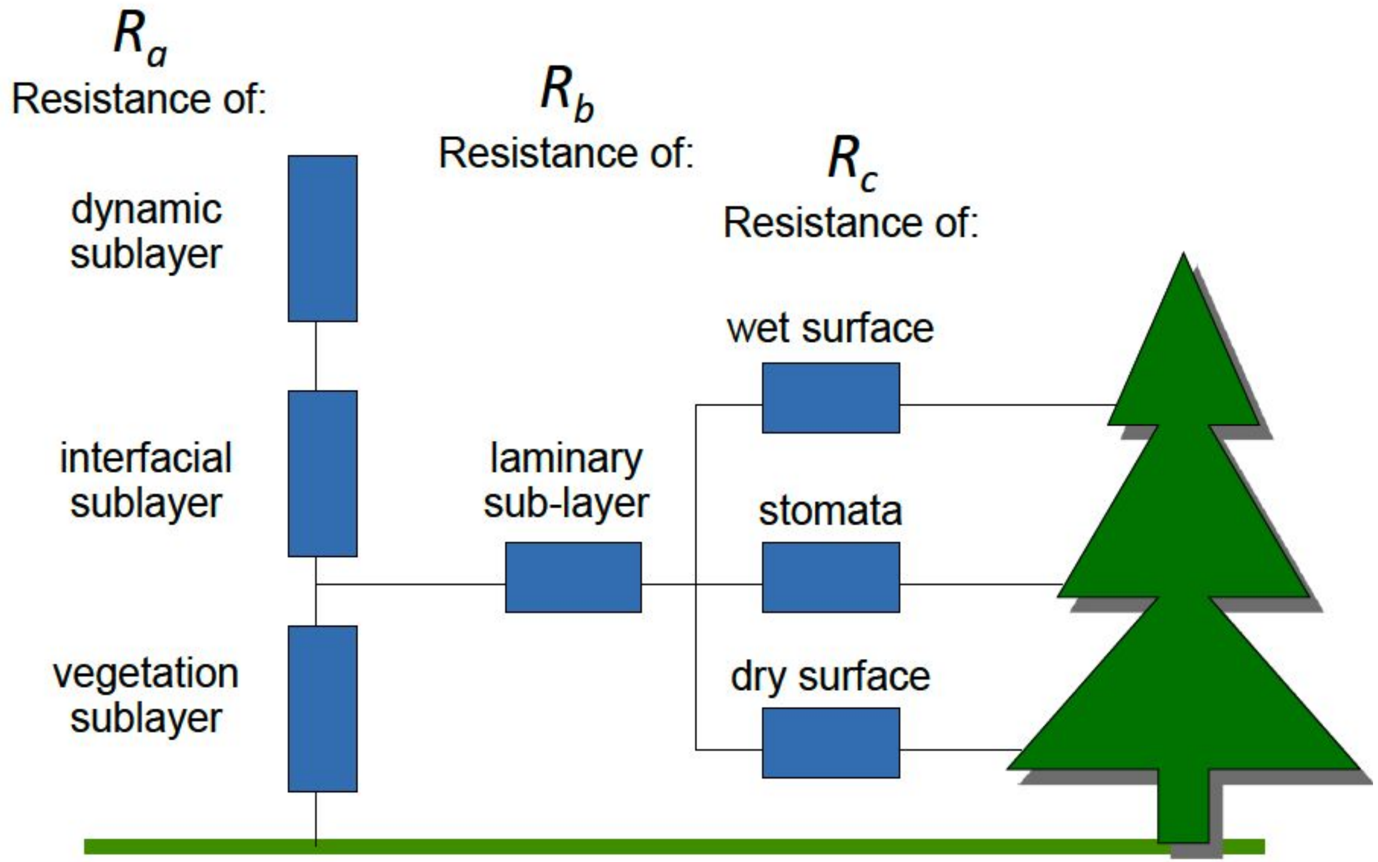
Resistance model:

$$V_d = \frac{1}{R_a + R_b + R_c}$$

$$\boxed{F = -v_d C}$$

F = deposition flux
C = concentration of species in 10m surface layer

Uptake of chemical constituents by plants and soil (CLM), depends on land type, roughness of surface



For each chemical constituent (χ), the following must be solved

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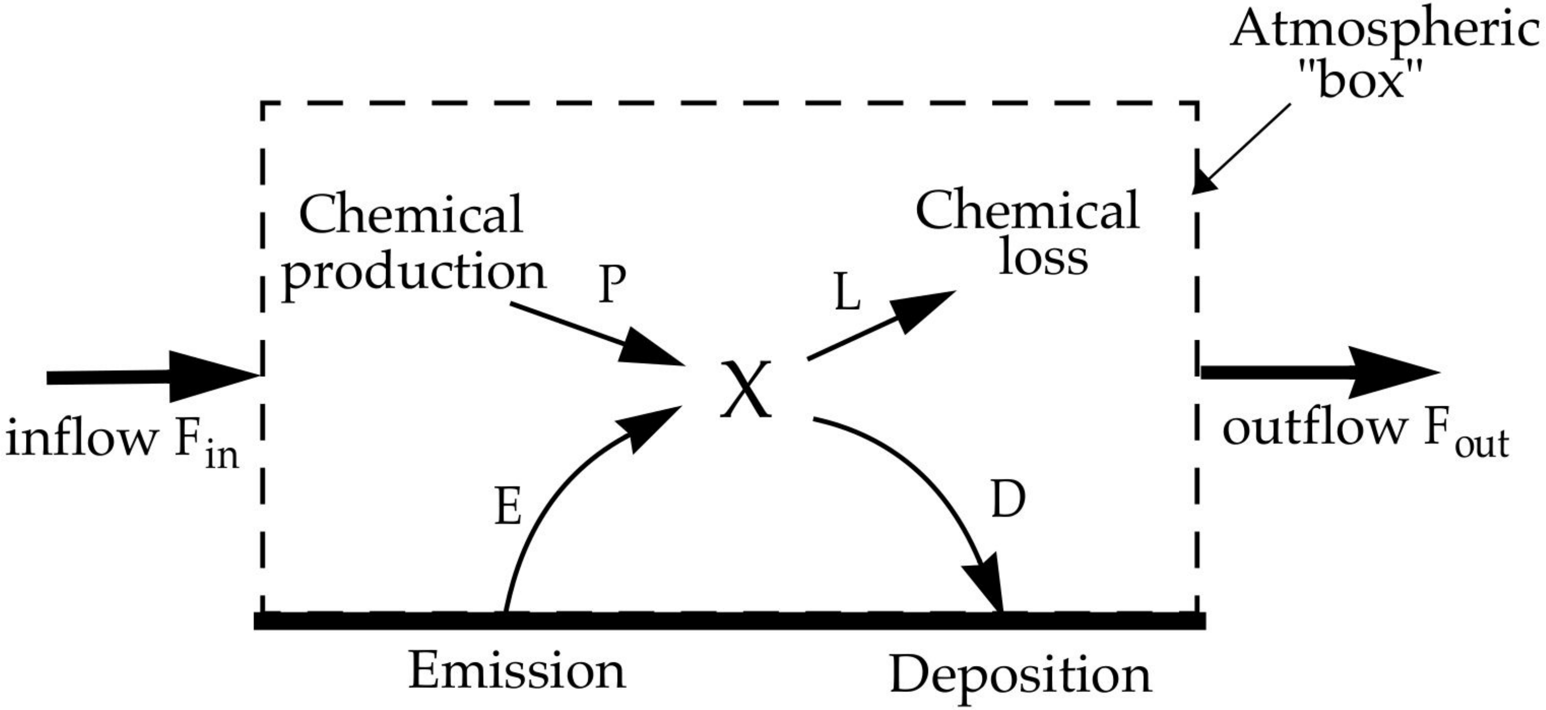
it can get expensive very fast! $

E_i	Emissions
C_i	Gas-phase-Chemistry
A_i	Aerosol-processes (Gas-aerosol exchange, het chem.)
T_i	Advection + Diffusion
W_i	Cloud-processes (wet deposition)
D_i	Dry deposition



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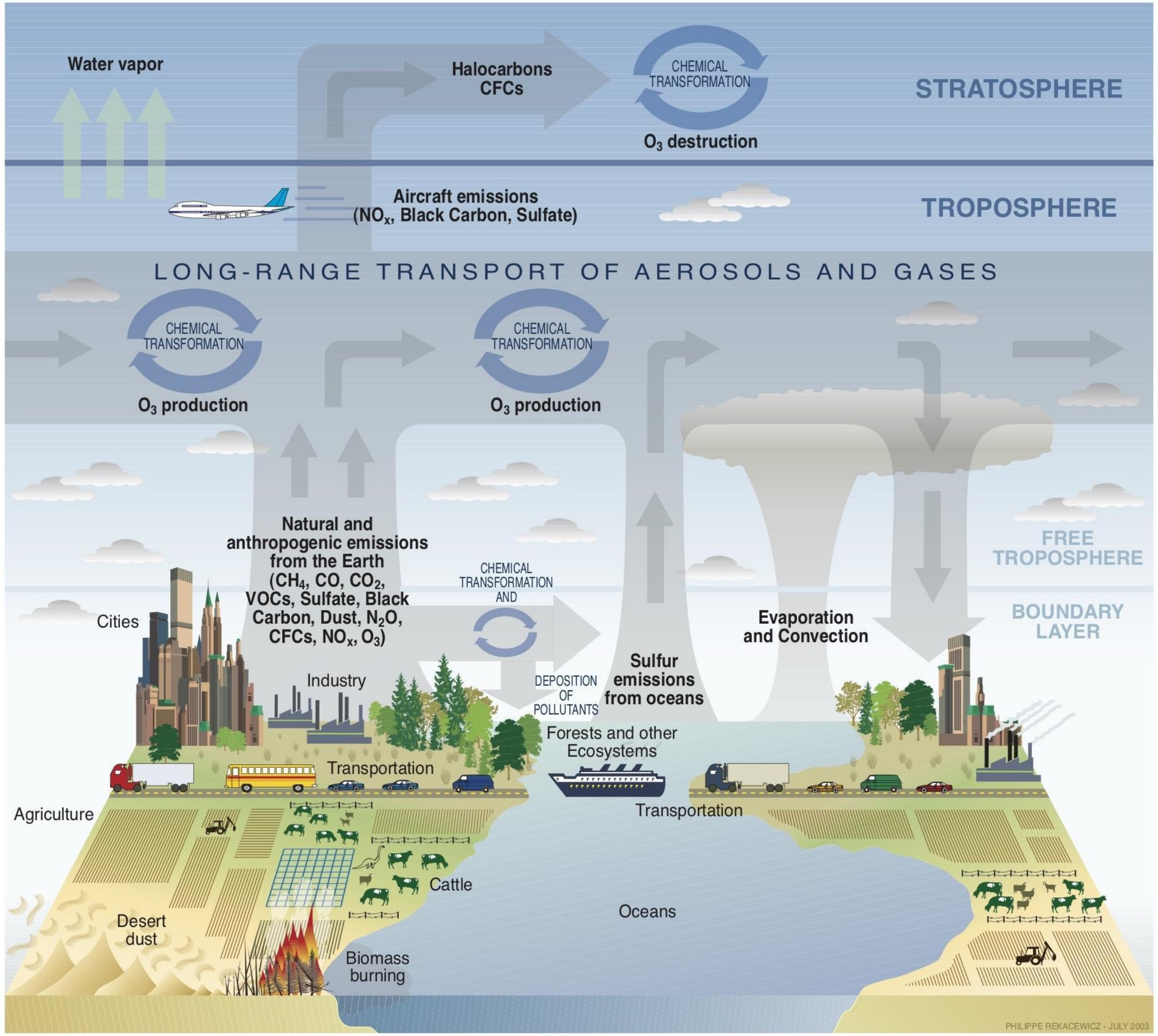
Atmospheric Chemistry

- Motivation
- Adding processes into models
  - Emissions
  - Chemical mechanism
  - Aerosol model and cloud interactions
  - Transport
  - Dry Deposition
  - Wet Deposition
- Applications: CESM
- Summary

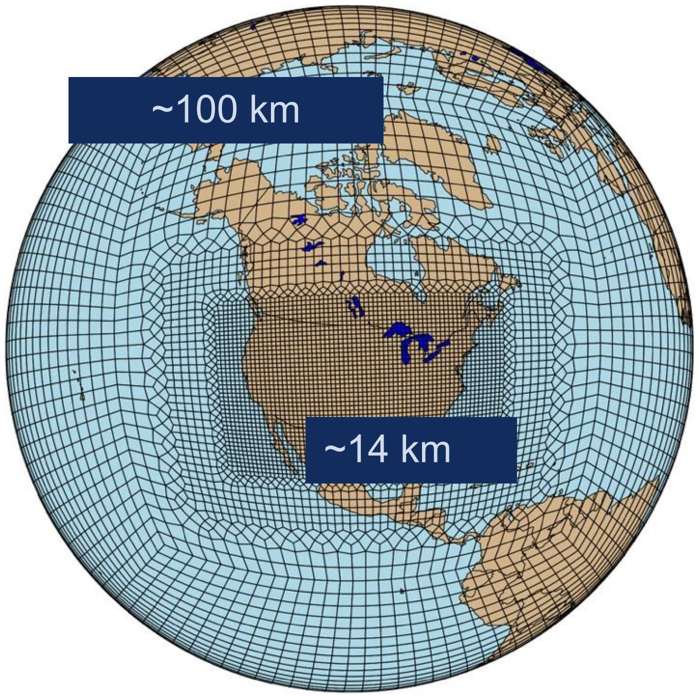


Chemistry → Air Quality: Regional refinement

MUSICAv0: **Multi-Scale Infrastructure for Chemistry and Aerosols**

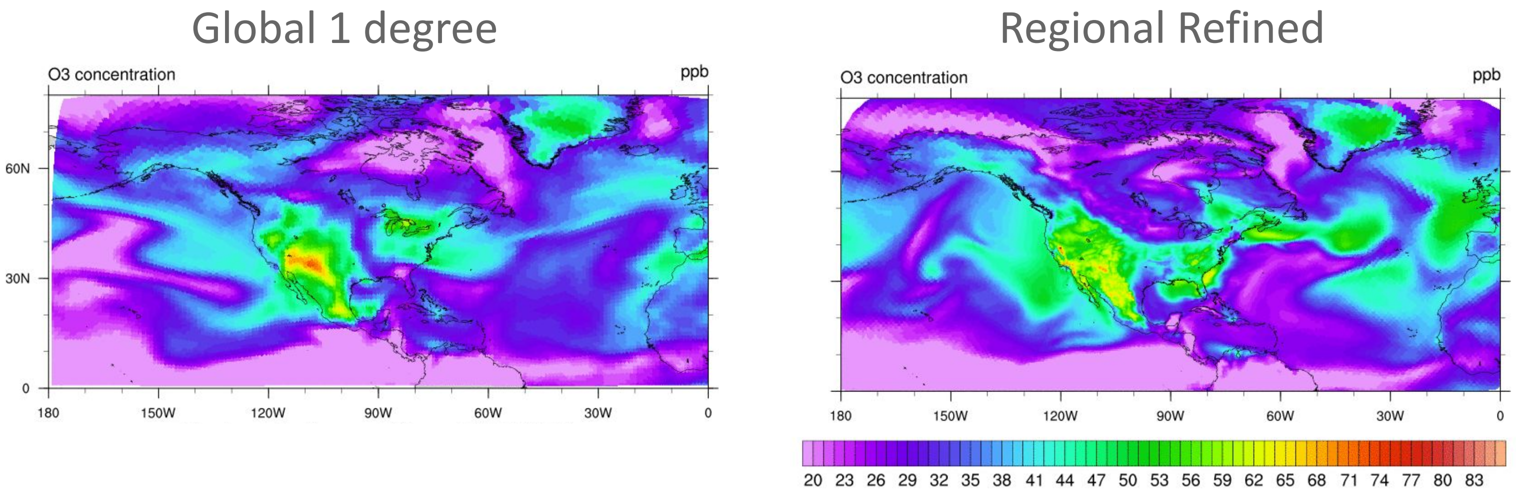
CAM-chem-SE-RR - Community Atmosphere Model with Chemistry With Spectral Element (SE) dynamical core and Regional Refinement (RR)

MUSICA-wiki: tutorials and support https://wiki.ucar.edu/display/MUSICA



Example: U.S. Air Quality, Surface Ozone (ppb)

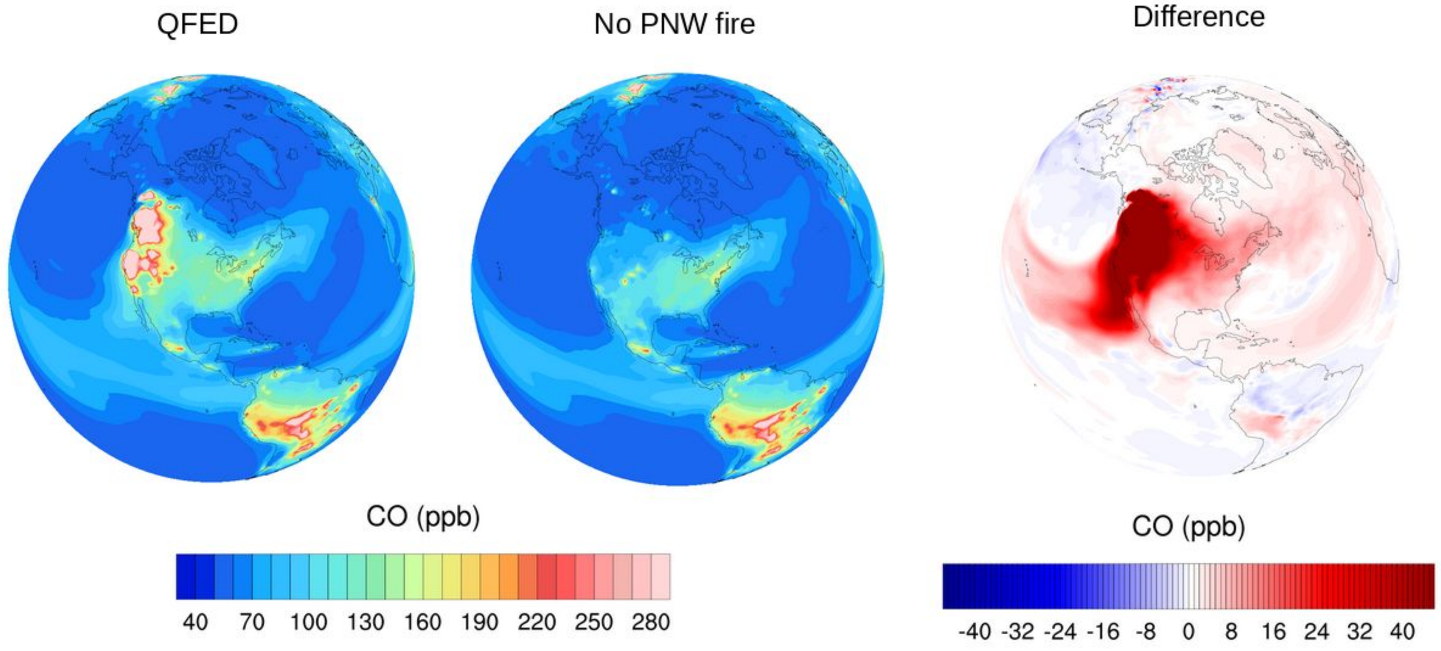
- Exposure Relevant scales and large-scale feedbacks



Credits: Forrest Lacey (ACOM)

Modeling potential wildfire impact on air quality

2018 August North American fire emissions in CAM-chem

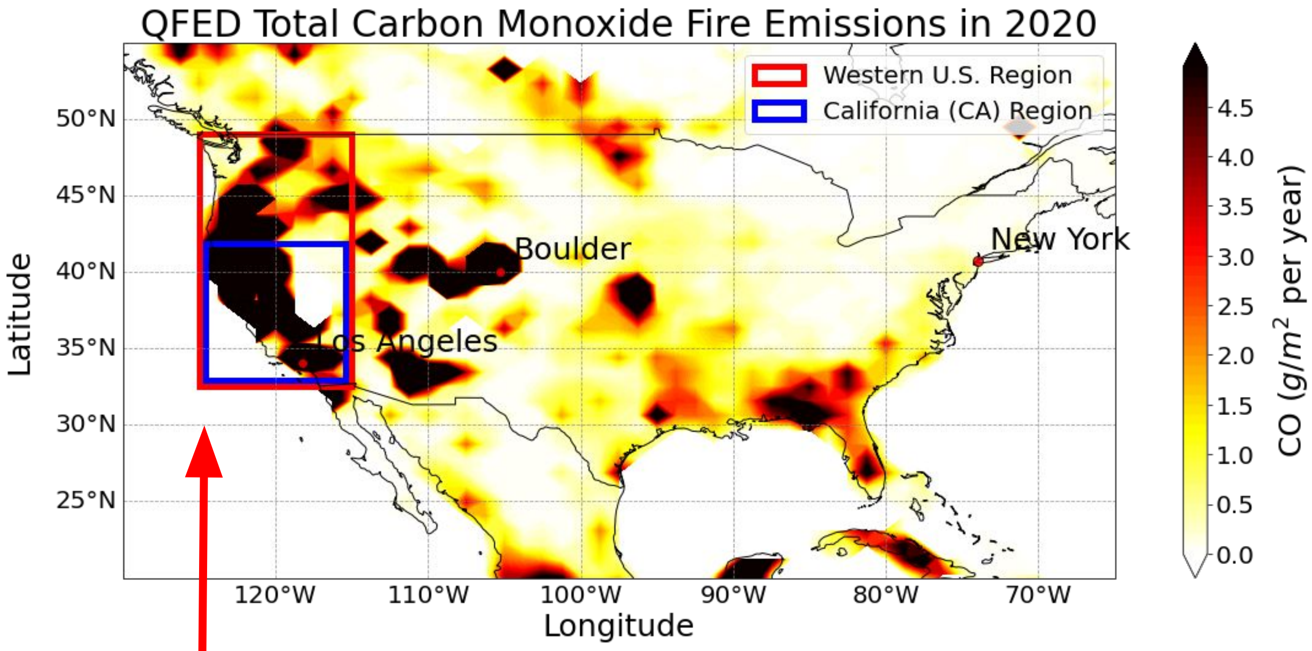


2020 ACOM-CU intern: Tom Sullivan

now: pilot

CAM-chem simulations with and without fire emissions in the Pacific Northwest (PNW) show impact on **downwind** atmospheric composition.

USA wildfires in 2020: Unprecedented emissions



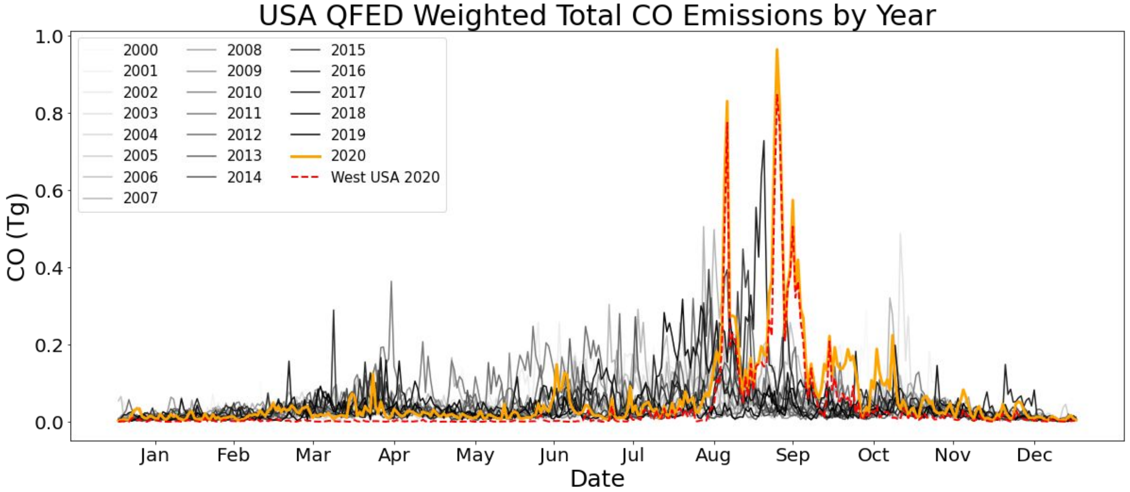
QFED Total Carbon Monoxide Fire Emissions in 2020

2021
SOARS Protegé:
Isabel Albores

now: PhD at MIT

Some regions in 2020 had >500% more emissions than the 2001-2019 average.

Fire emissions started and ended later than in previous years.

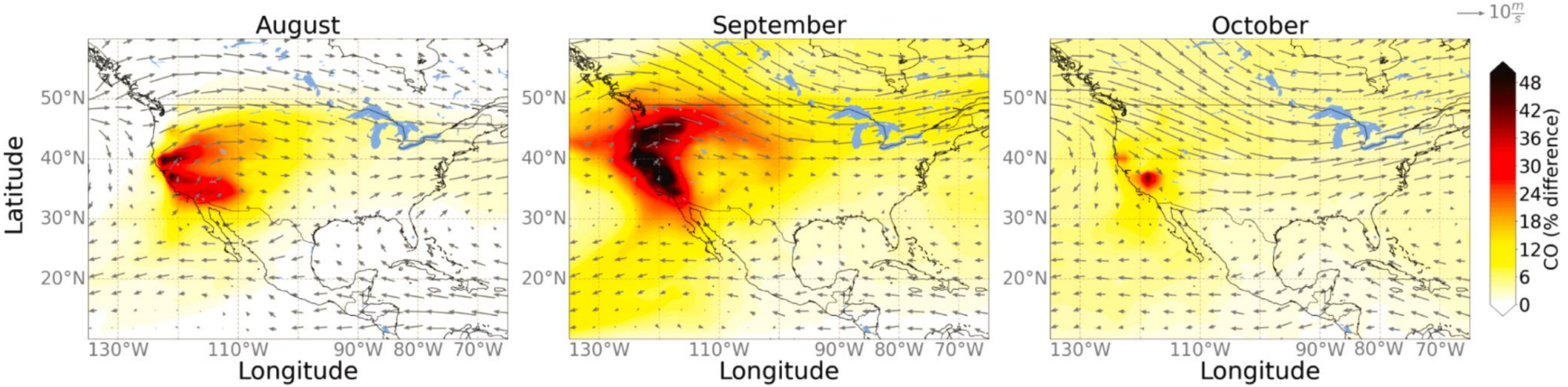


USA QFED Weighted Total CO Emissions by Year

Albores et al., Atmos. Environ., 2023

USA wildfires in 2020: Column CO differences

CESM/CAM-chem simulations with and without fire emissions in the Western U.S. show impact on **downwind** atmospheric composition

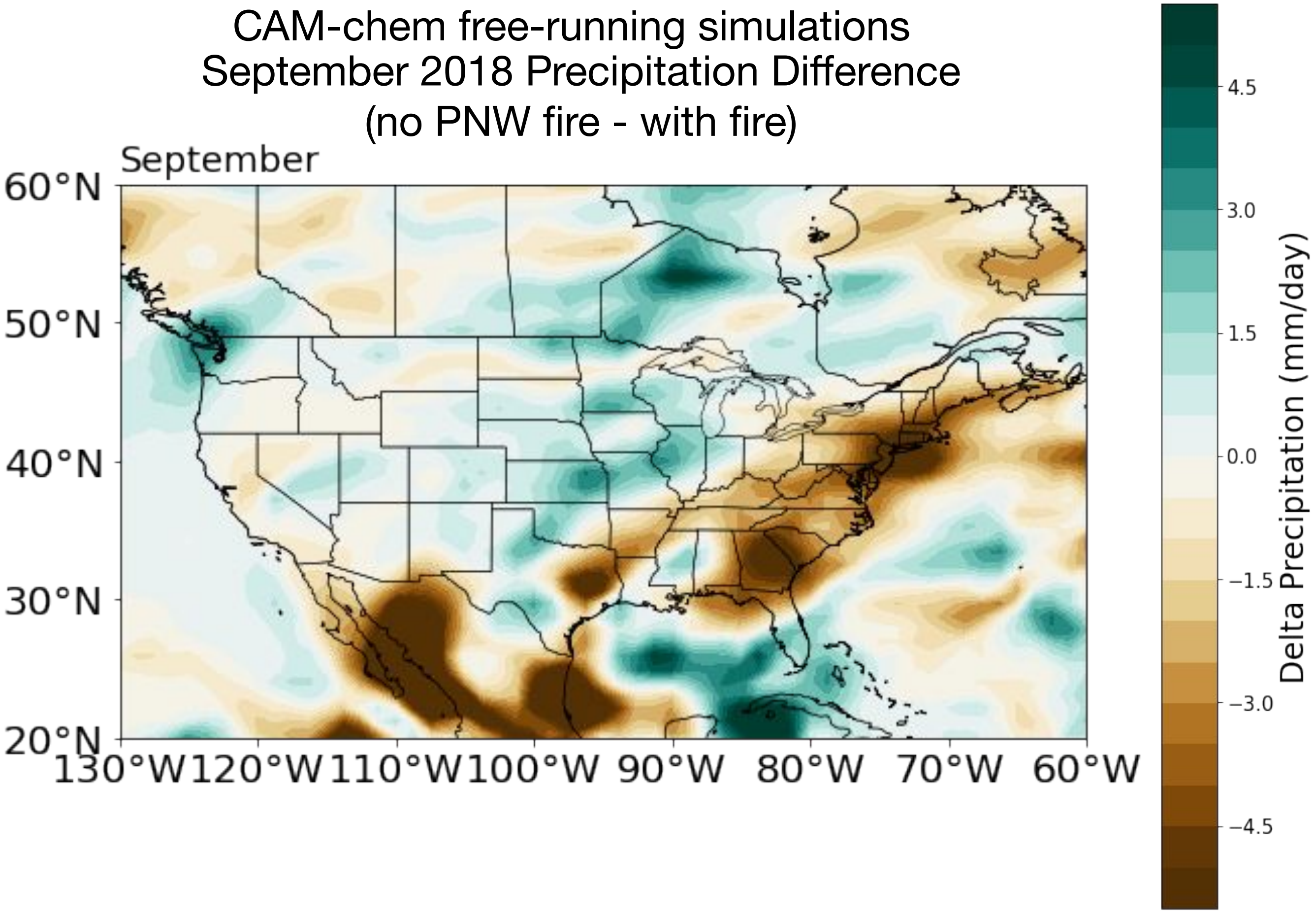


CONUS impact: 6.5 % 14.5% 4.9%

Albores et al., Atmos. Environ., 2023

Chemistry → Weather: Pacific Northwest (PNW) wildfire emissions impact on precipitation

CAM-chem free-running simulations
September 2018 Precipitation Difference
(no PNW fire - with fire)



ACOM-CU intern:
Peizhi Hao

September, 2018 East Coast **precipitation decreased** when PNW wildfire emissions were turned off.

Impacts on precipitation occur via cloud microphysics (e.g. cloud fraction), and atmospheric dynamics (e.g. the 250 mb Jet Stream).

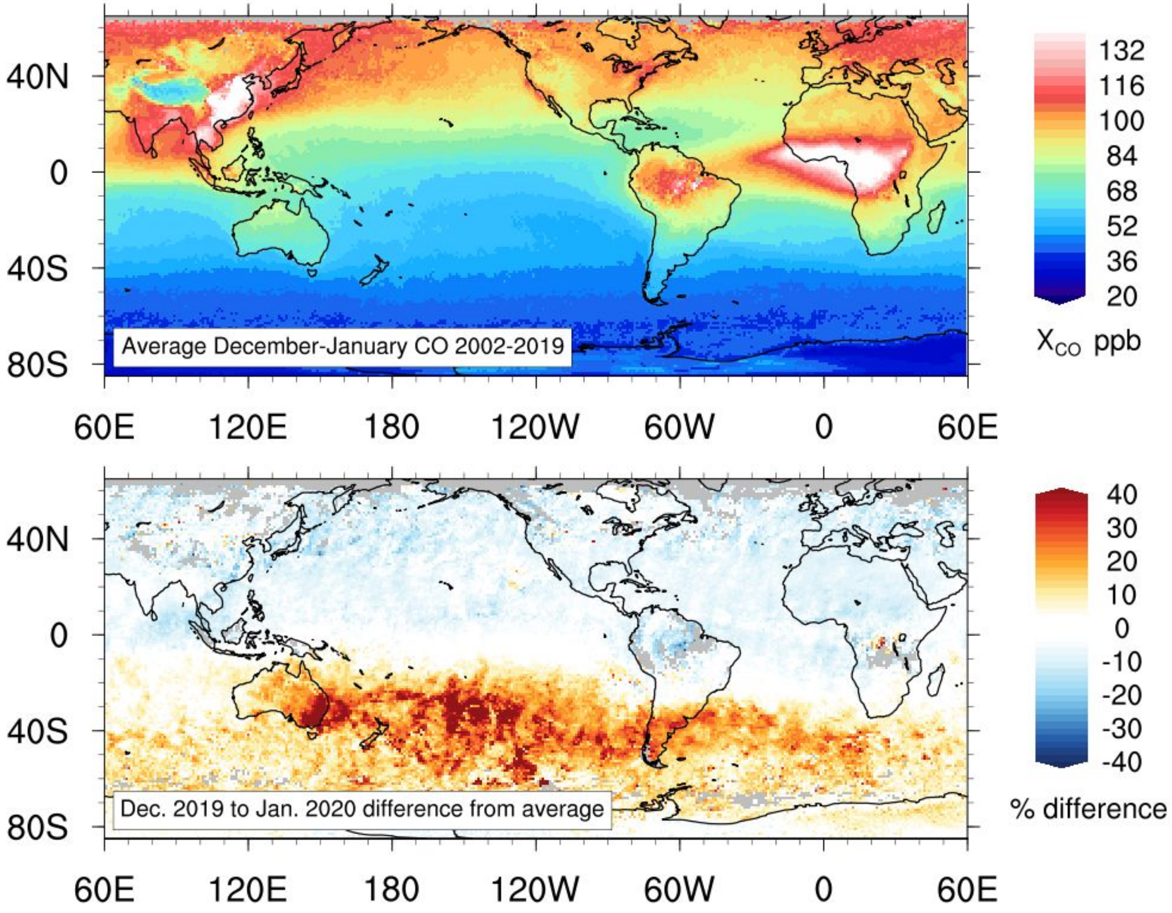
Australia: on fire from July 2019 to March 2020

Chemistry → Climate: Australian wildfires 2019/2020

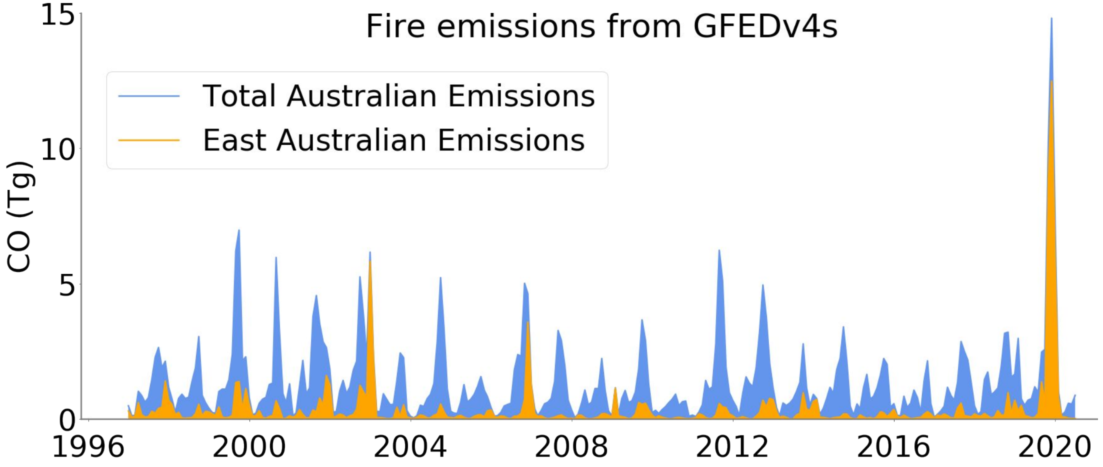
Impacts on:

- Ocean biogeochemistry - CO_2 offset (https://www.nature.com/articles/s41586-021-03805-8)
- “Caramelized” New Zealand glaciers (https://www.cnn.com/2020/01/02/australia/new-zealand-glaciers-australia-bushfire-intl-scli)
- Stratosphere: mid-lat ozone depletion (https://doi.org/10.1073/pnas.2117325119)
- NH/SH imbalance and ENSO (https://doi.org/10.1029/2021GL093841; https://doi.org/10.1126/sciadv.adg1213)

Satellite measured carbon monoxide

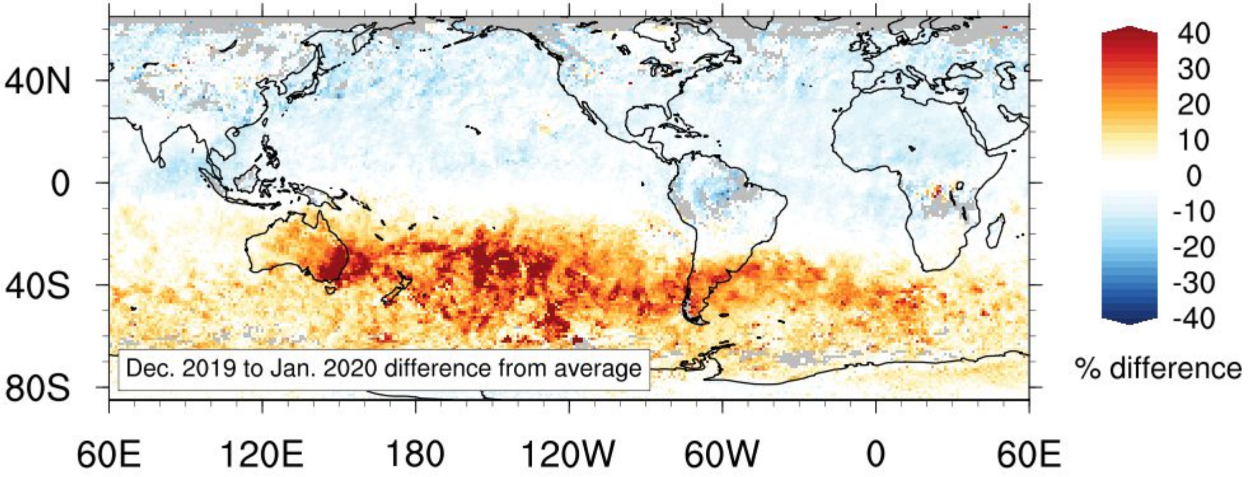


Chemistry → Climate: Australian wildfires 2019/2020

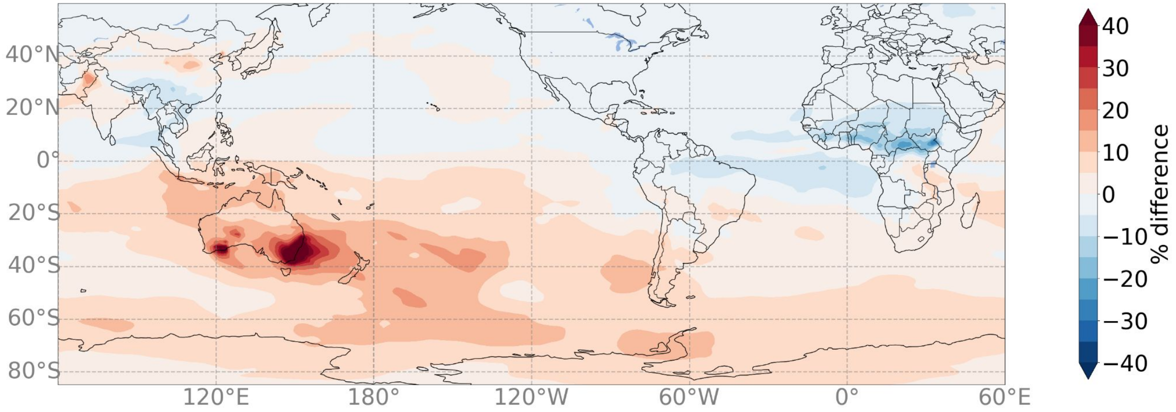


- CESM reproduces a similar magnitude response for the 2019/2020 difference in CO, compared to the rest of the record.
- Can be used to investigate the impacts of the extreme fire season on the Earth System.

Measurements: MOPITT



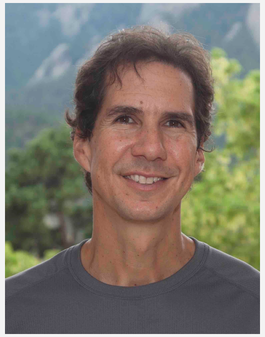
Model: CESM/CAM-chem



https://doi.org/10.5065/XS0R-QE86

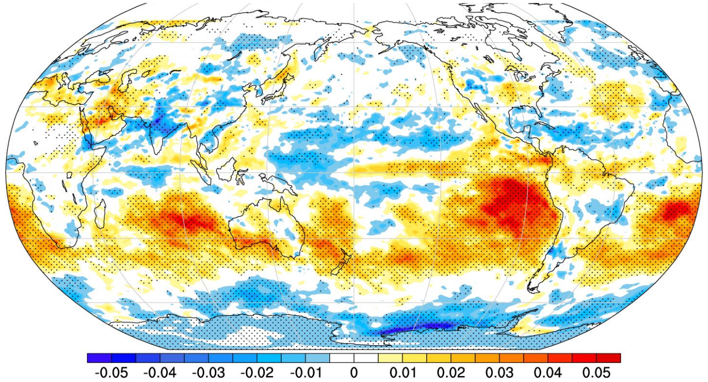
Chemistry → Climate: Australian wildfires 2019/2020

- CESM/CAM6 simulation with aerosols, satellite-based inventory (GFED) in Australia compared to climatology
- Climate response similar to a major volcanic eruption (aerosol-cloud interactions)
- Large interhemispheric radiative imbalance anomaly and impacts on ENSO



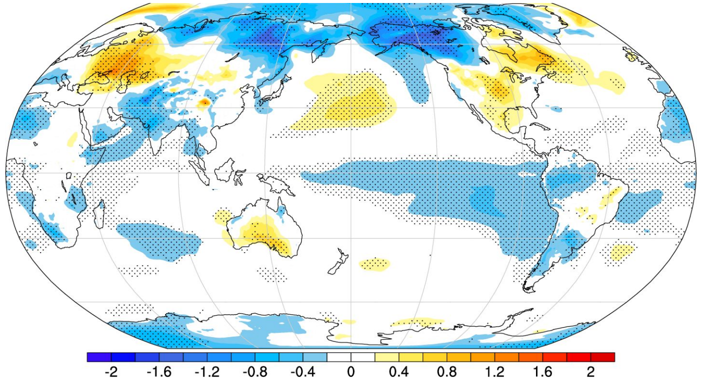
John Fasullo, CGD

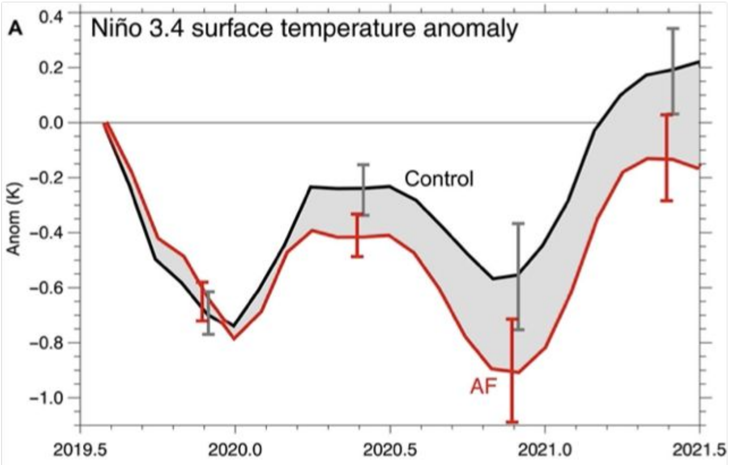
Cloudy Sky Albedo (Jan 2020)



Cloud brightening across the Southern Hemisphere

Near-surface temperature (Jan 2021)





2020/21 La Niña response

Fasullo et al., GRL, 2021

Fasullo et al., Sci. Adv., 2023

Atmospheric Chemistry

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- Applications
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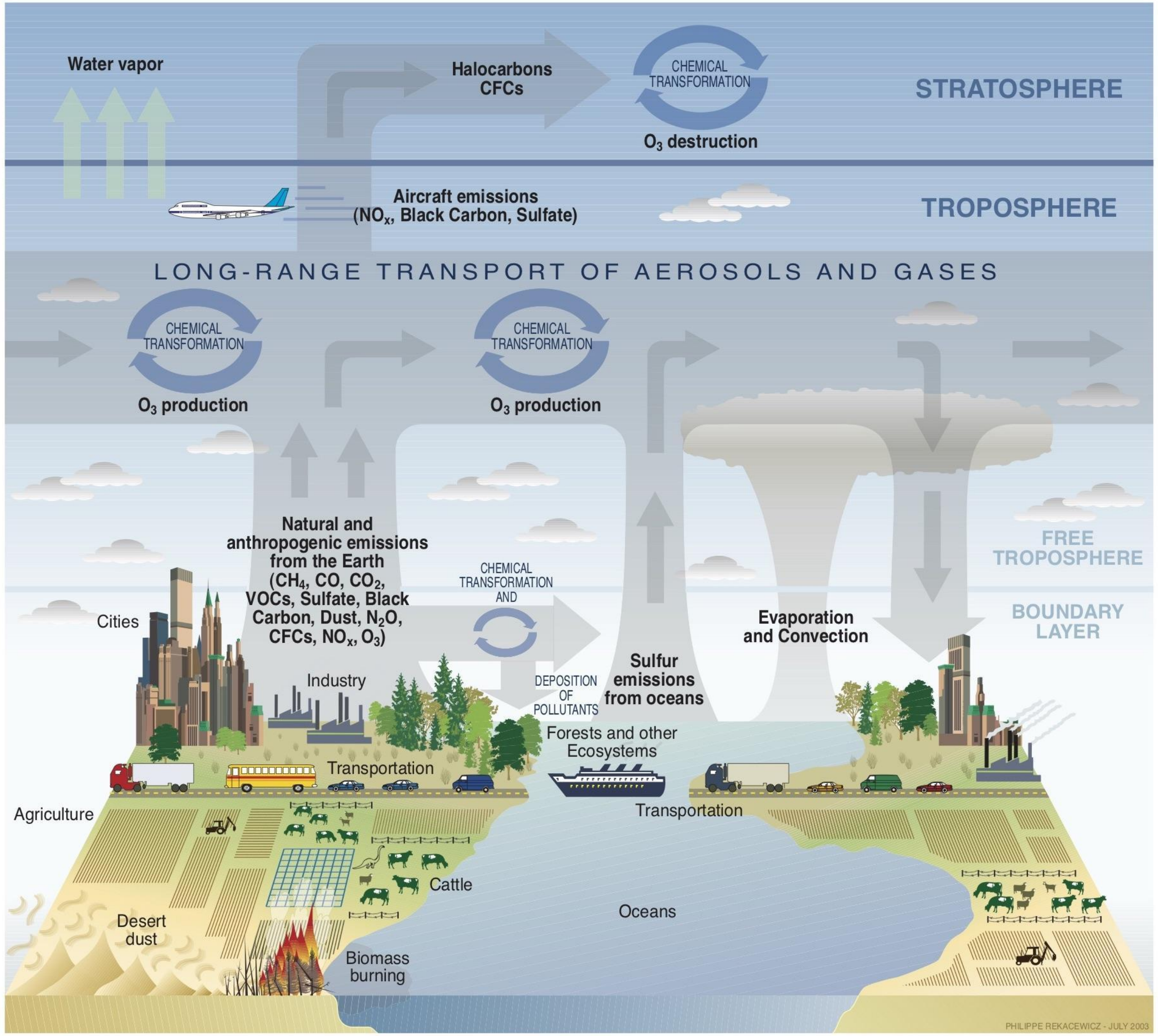

Key takeaways

- Atmospheric chemistry is important in models due to the feedback into the earth system. It has **impacts on health, weather and climate**.

- Adding atmospheric chemistry processes into earth system models requires many **approximations and parametrizations**

$$\boxed{\frac{\partial \chi(i)}{\partial t} = \mathrm{Sources(i)} - \mathrm{Sinks(i)} = \mathrm{E_i} + \mathrm{C_i} + \mathrm{A_i} + \mathrm{T_i} - \mathrm{W_i} - \mathrm{D_i}}$$

    - Considerations include: Emissions, Chemical mechanism, Aerosol model and cloud interactions, Transport, Dry Deposition, Wet Deposition

- Models allow us to perform multiple experiments regarding our atmosphere. Using the correct model or model configuration is important to correctly answer your question.