Modeling Atmospheric Chemistry and Aerosols

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Shawn Honomichl

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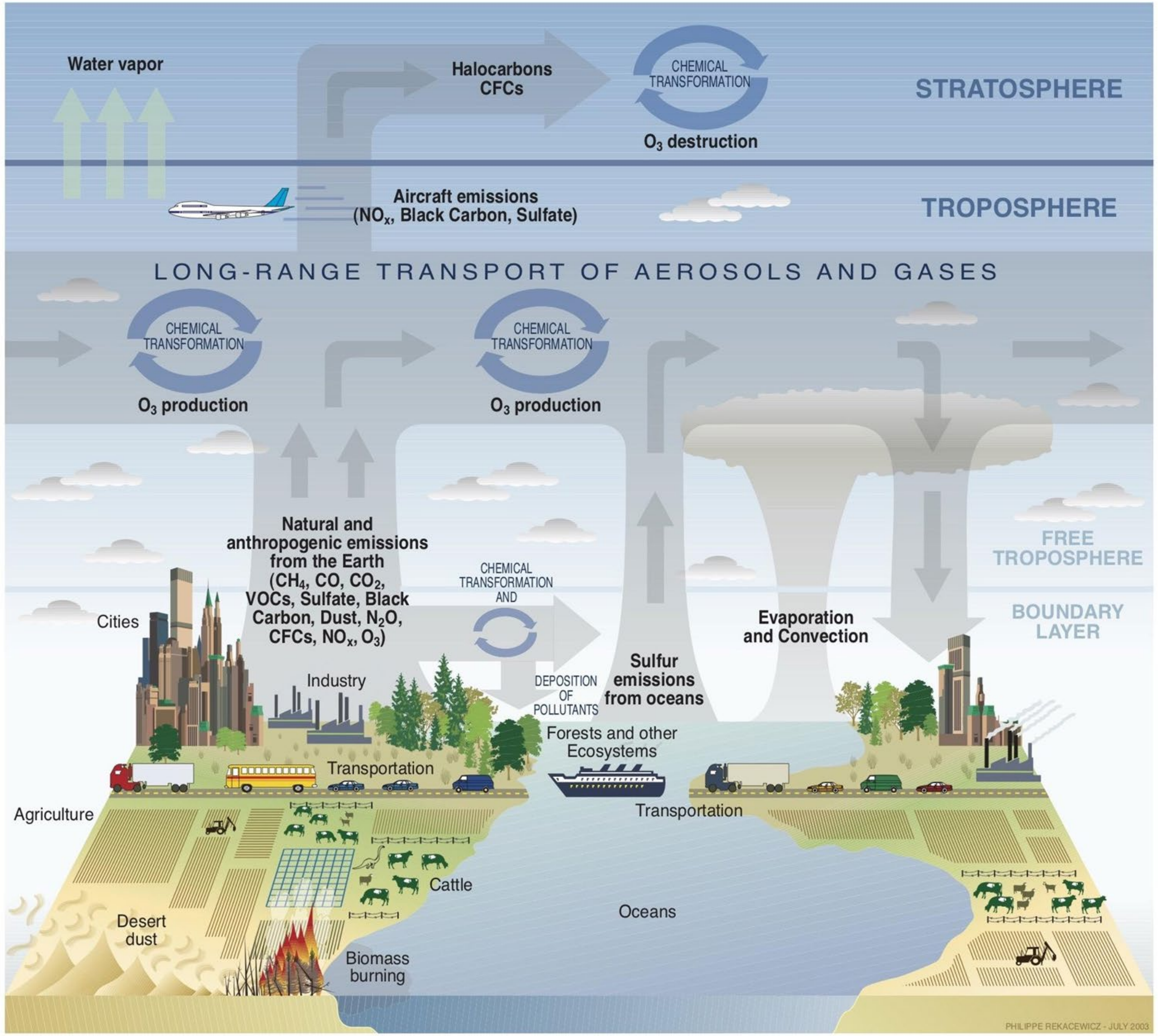
Atmospheric Chemistry

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



Atmospheric Chemistry: Why is it important – Health

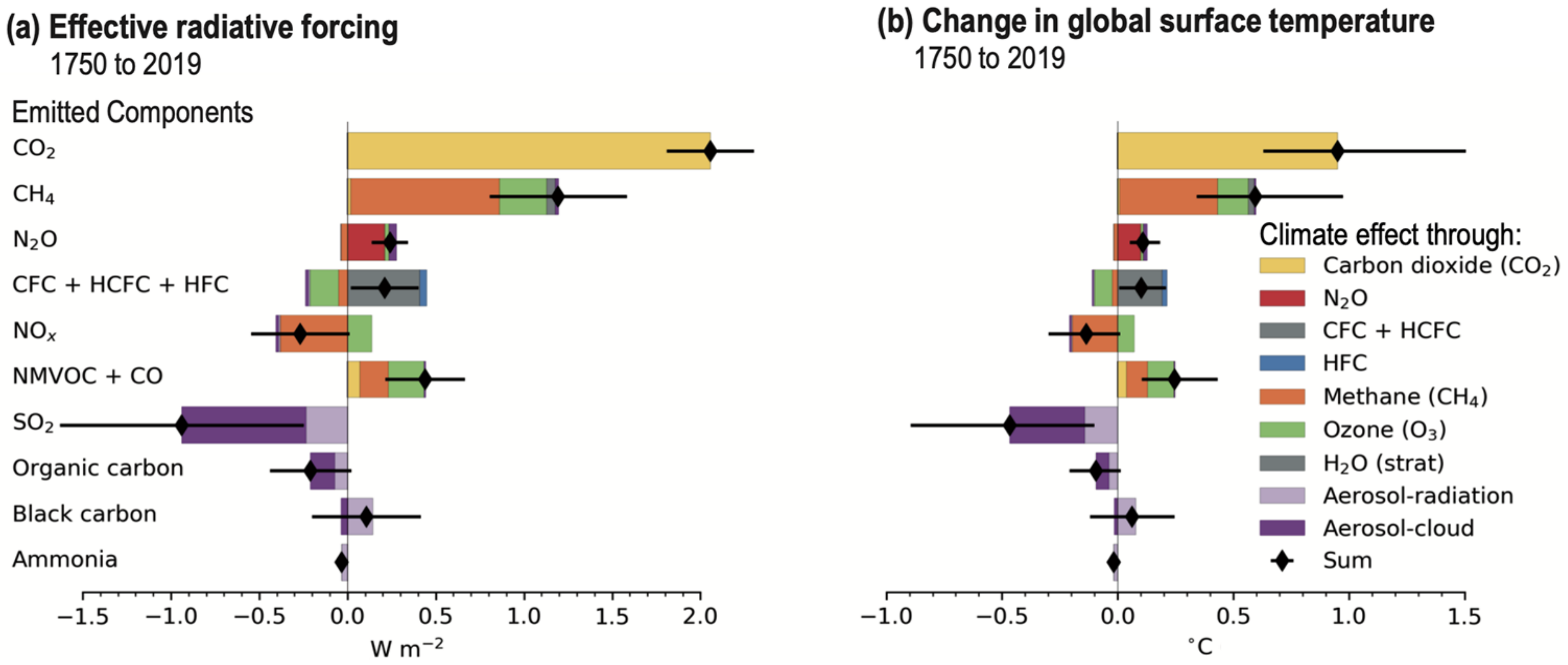
Ozone pollution (NOx, CO, VOC, CH_4):
→ Damages tissues, causes inflammation
→ Coughing, chest tightness and worsening of asthma

Particulate Matter: PM2.5 and PM10 diameter < 2.5 or 10 µm
(SO_2, VOC, NH_3, 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)

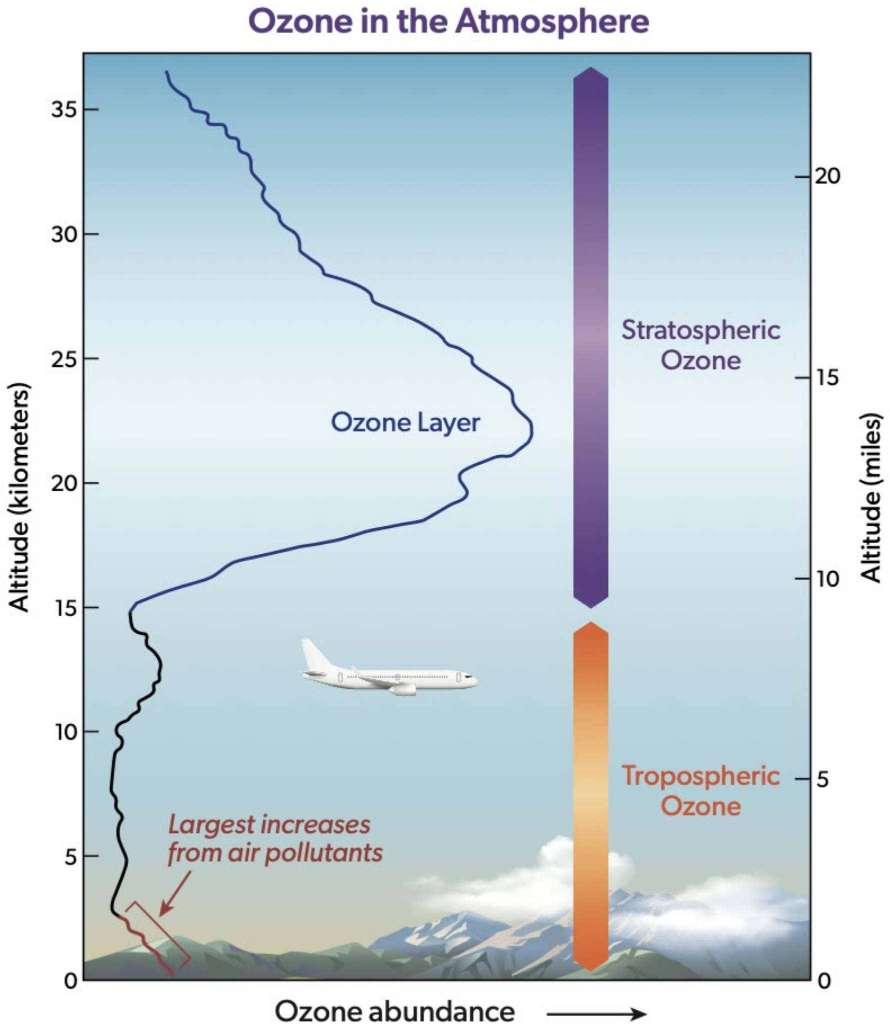
Atmospheric Chemistry: Why is it important – Climate

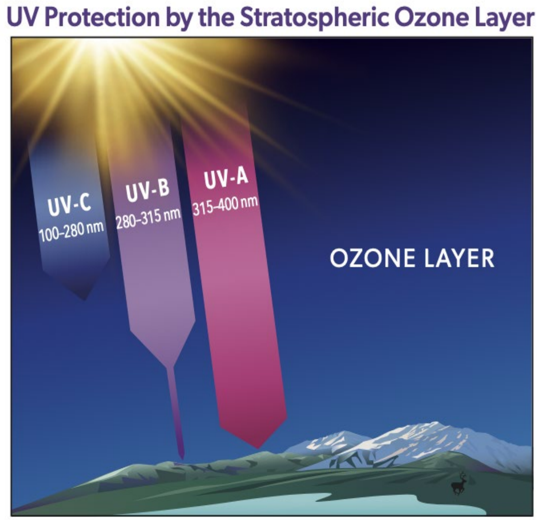


IPCC AR6 WG1 Technical Report, Figure TS15

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

Atmospheric Chemistry: Why 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)
- e.g. cause of a slowing trend

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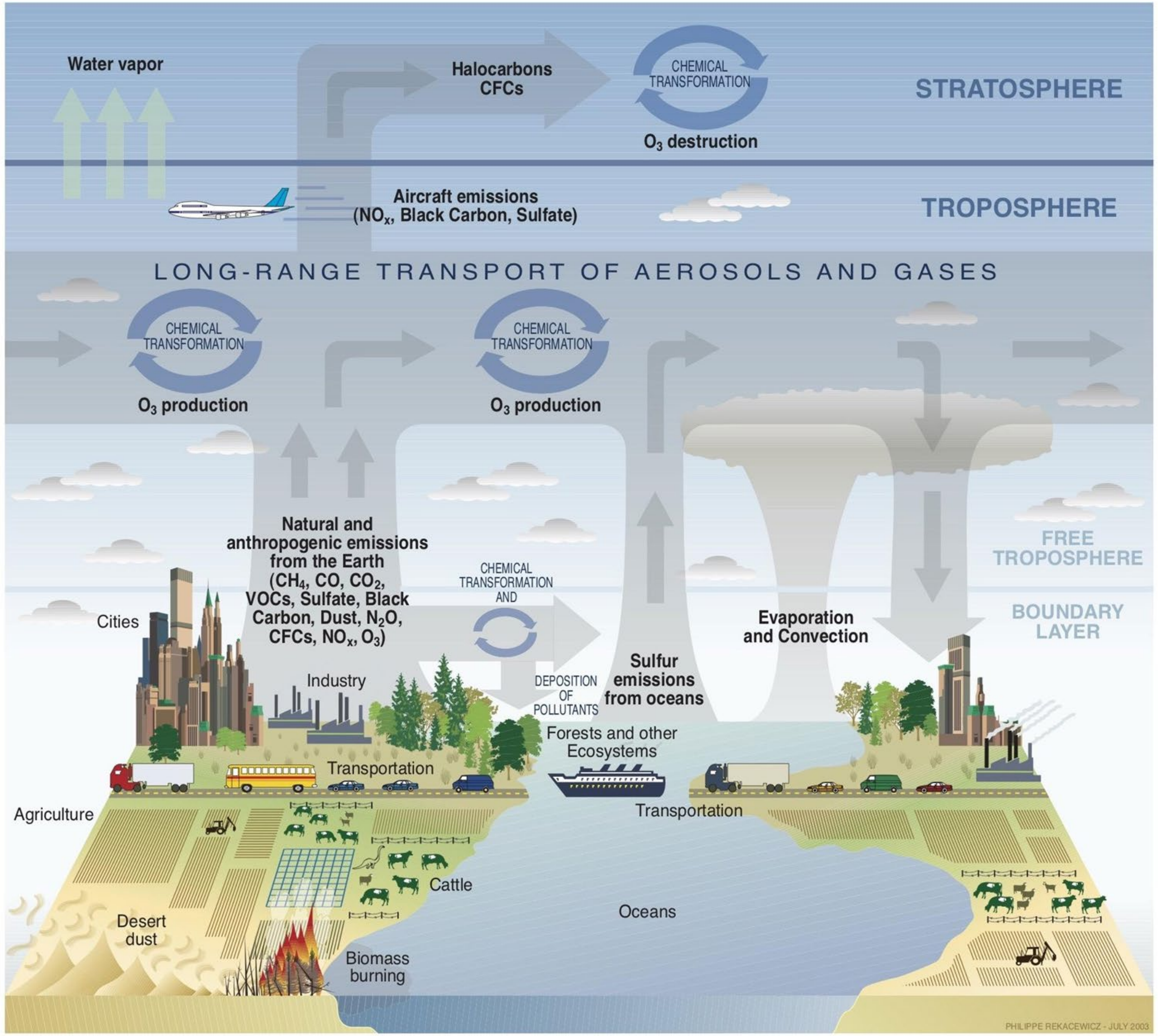

For each chemical constituent (χ), the following must be solved

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

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

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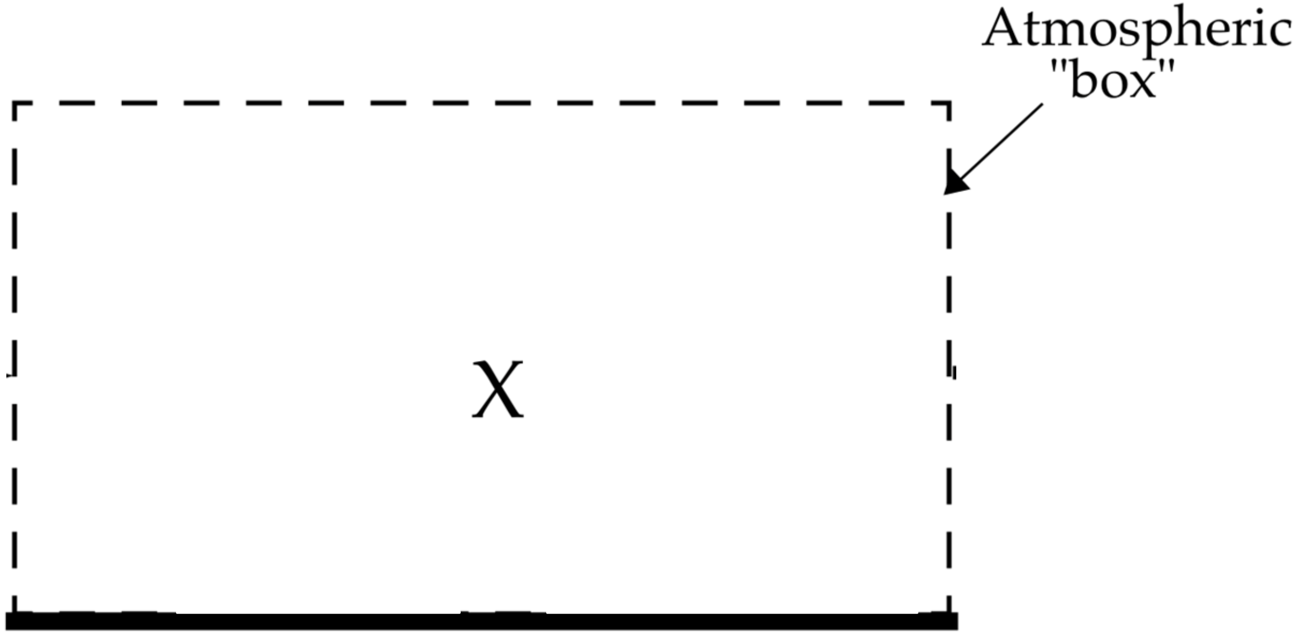

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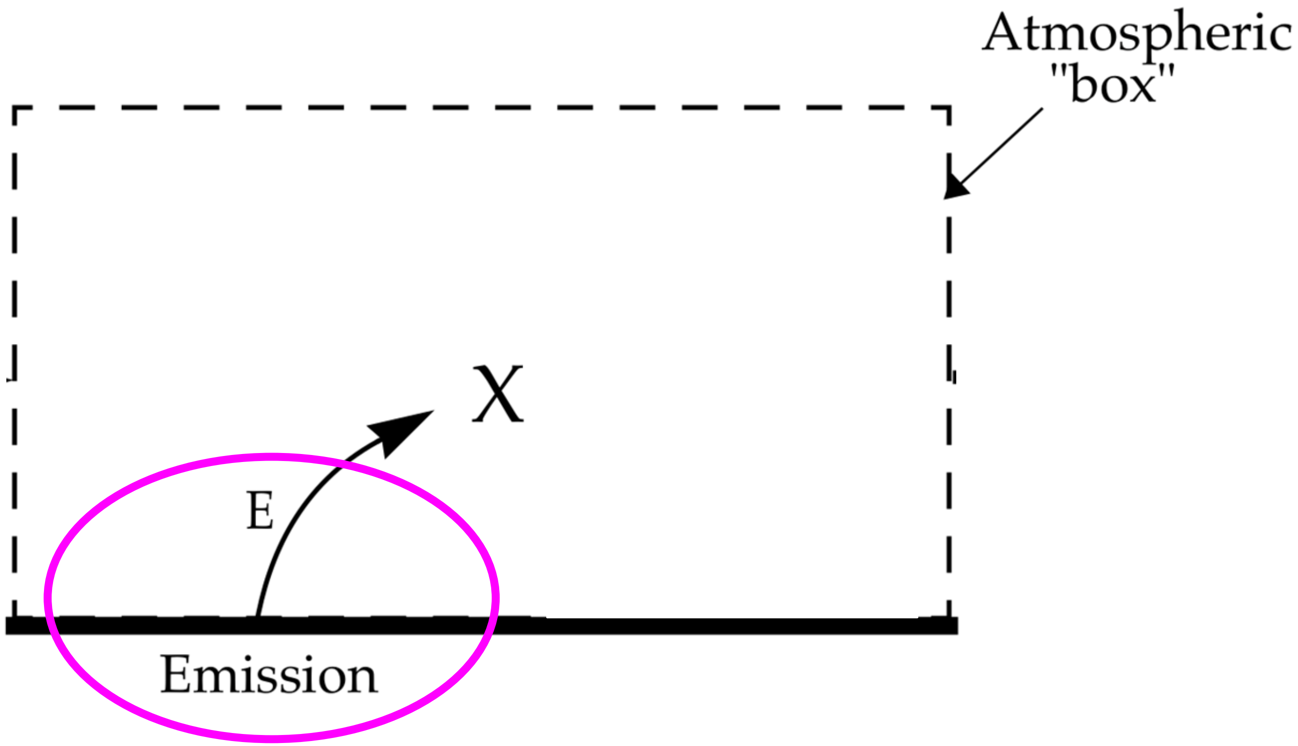
E_i Emissions



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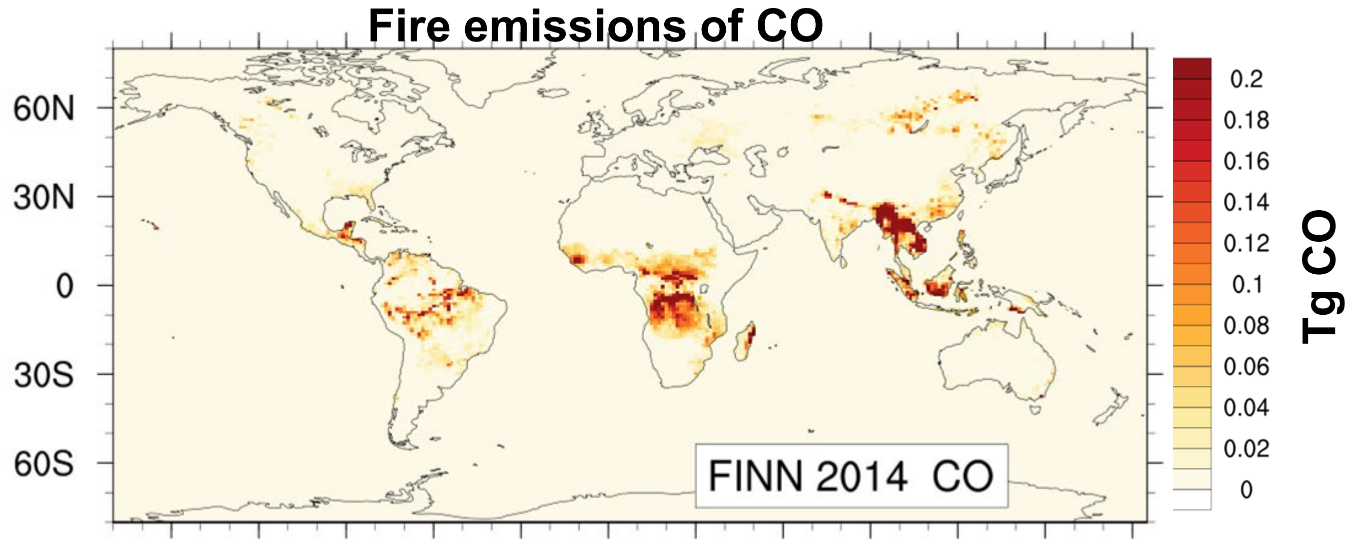
Emissions in CESM: 4 main "types"

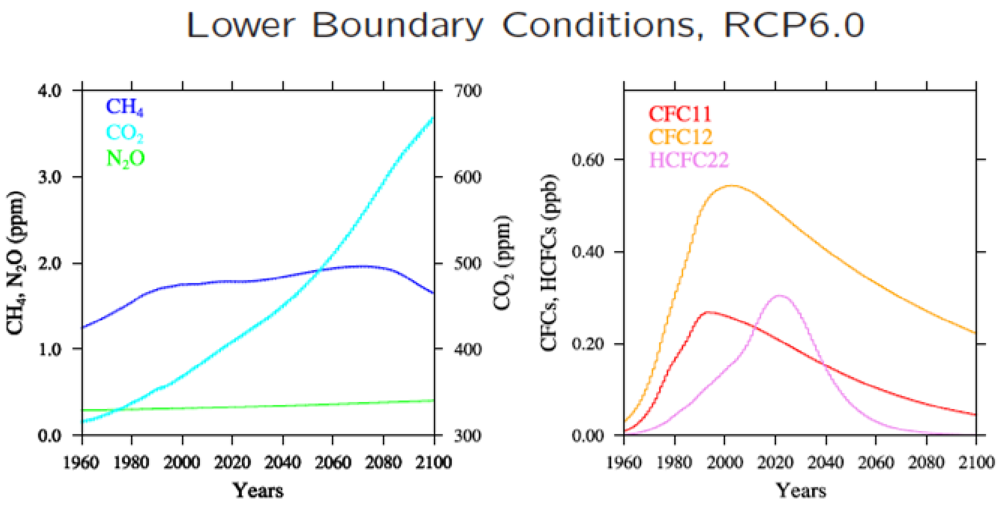
Emissions

- 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.





Interactive emissions: Dust

$F_{dustemis} = F_{dustemis}(u_*, w)$ becomes $F_{dustemis} = F_{dustemis}(u_*, w, z_{0,rock}, LAI, \sigma_{\tilde{u}})$

(CESM default)

horizontal wind/friction (u_*)

soil moisture (w)

(Leung 2023)

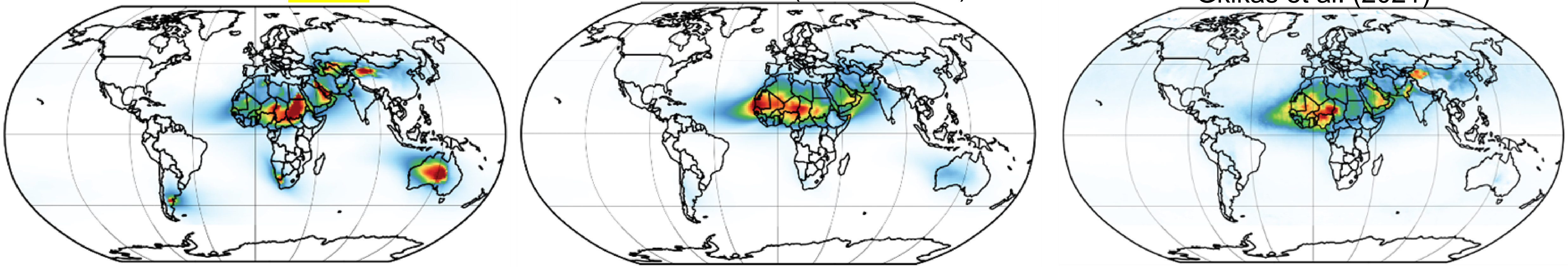
Drag partition due to surface roughness ($z_{0,rock}$, LAI)

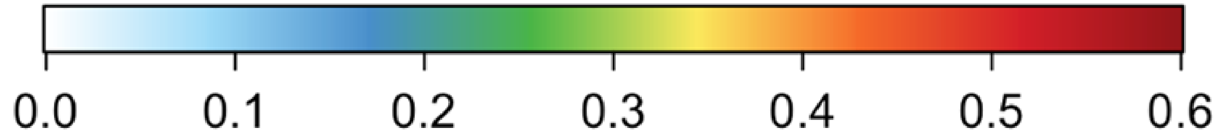
Subtimestep wind following the similarity theory ($\sigma_{\tilde{u}}$)

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)





Slide: Danny Leung

Interactive emissions: Biogenic

The **MEGAN-v2.1** algorithm
Emissions for species i:

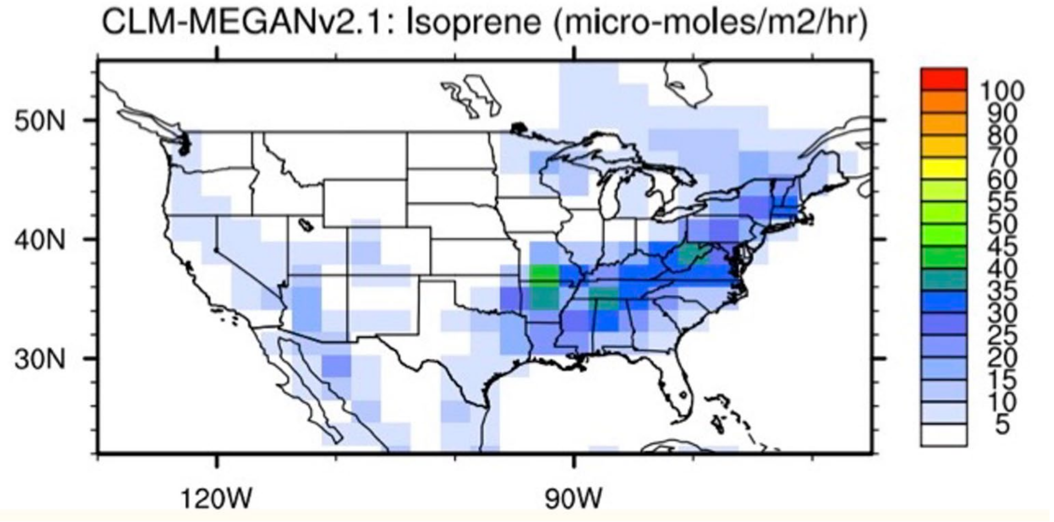
$$F_i = \gamma_i \; \Sigma \, \varepsilon_{i,j} \, \chi_j$$

where

γ_i : emission activity factor, depends on leaf area index (LAI), meteorology (T, solar radiation), leaf age, soil moisture, 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



Guenther et al., GMD, 2012

Slide: Louisa Emmons

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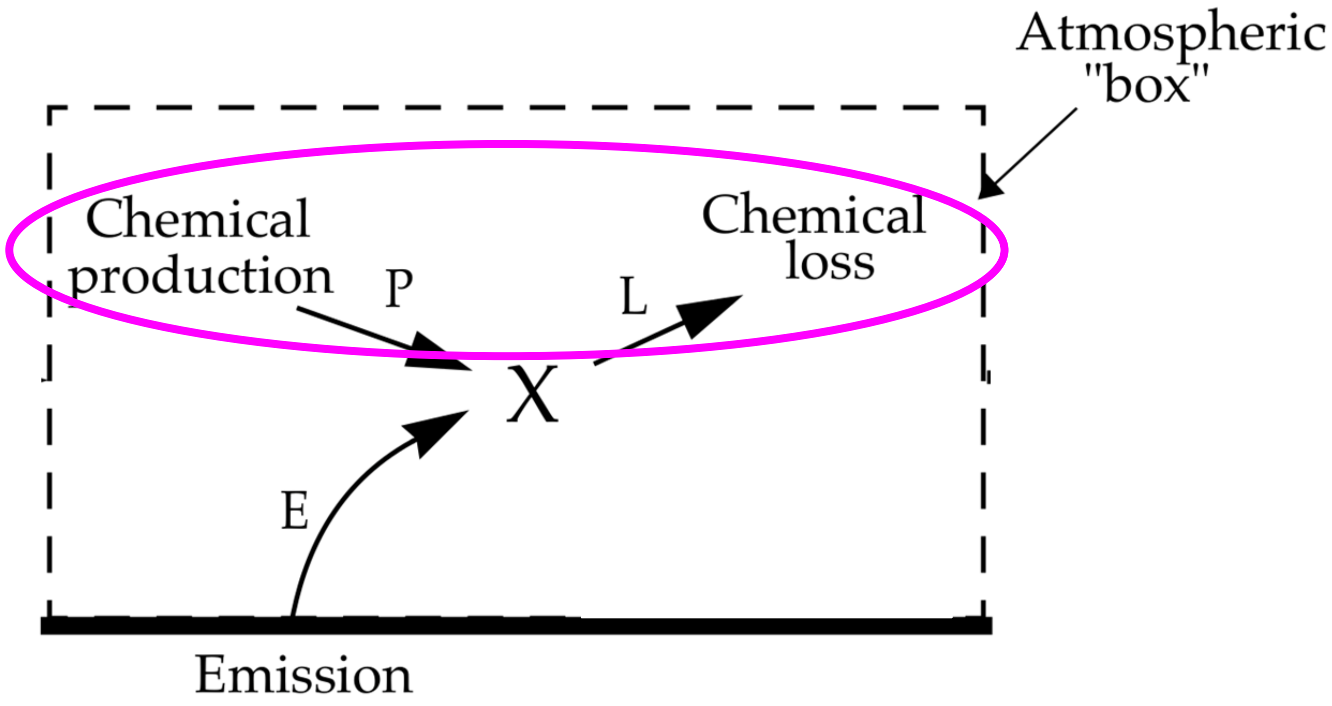
E_i Emissions

C_i Gas-phase-Chemistry

A_i Aerosol-processes (Gas-aerosol exchange, het chem.)



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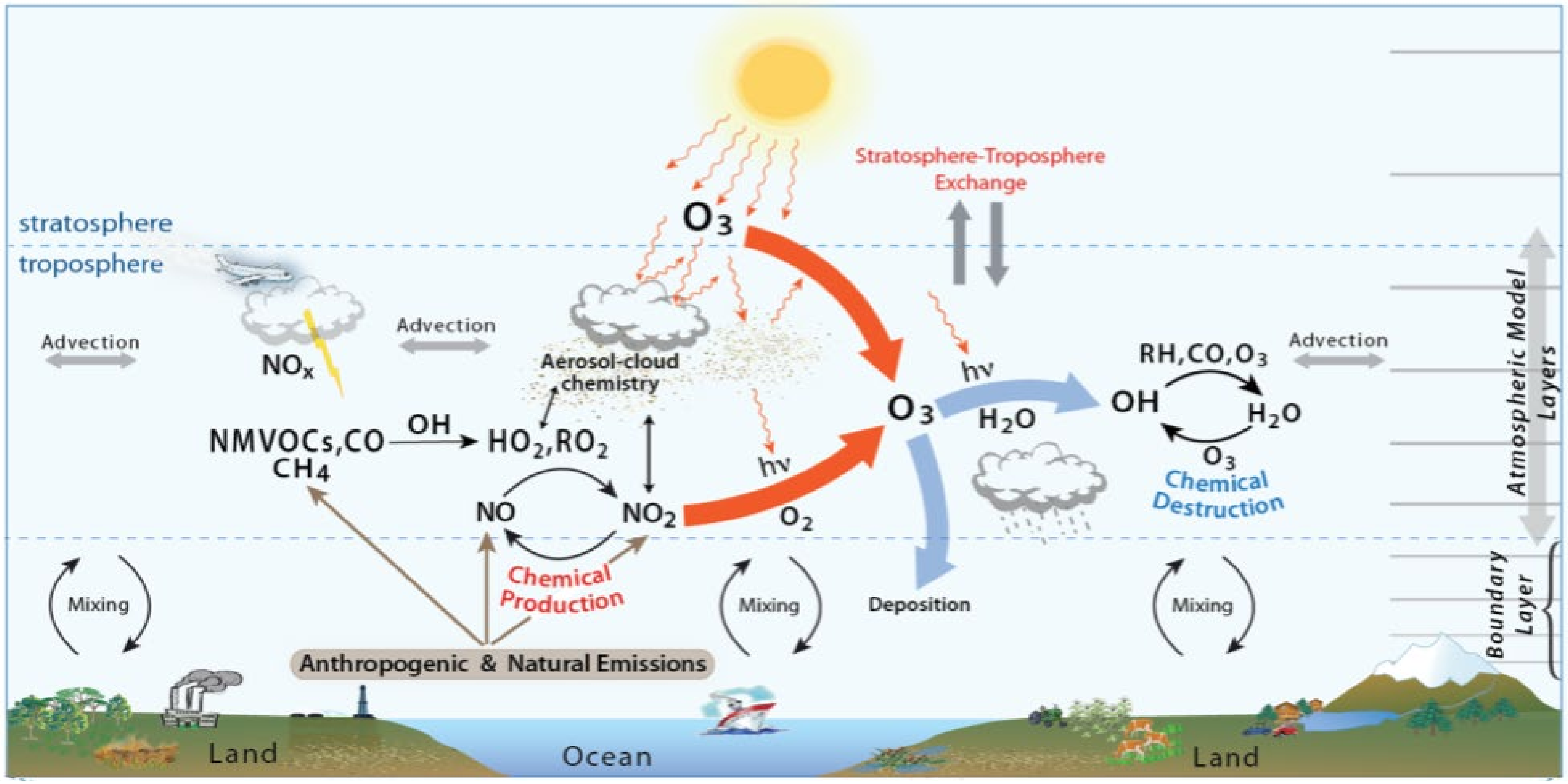
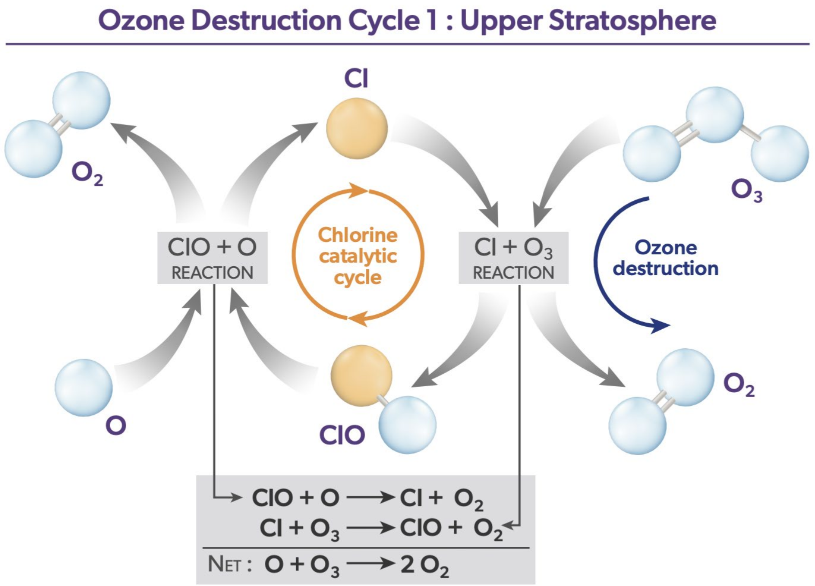
Tropospheric Chemistry



Young et al., 2018, https://doi.org/10.1525/elementa.265

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

Stratospheric Chemistry



WMO2022

Ozone Destruction Cycles 2 and 3 : Polar Regions

CYCLE 2 :

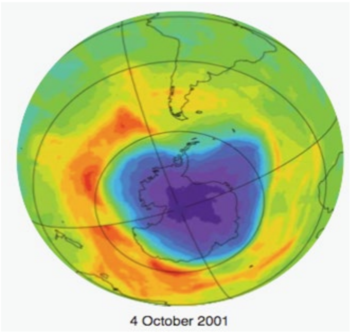
$$\mathrm{ClO + ClO \longrightarrow (ClO)_2}$$
$$\mathrm{(ClO)_2} + \textit{sunlight} \longrightarrow \mathrm{ClOO + Cl}$$
$$\mathrm{ClOO \longrightarrow Cl + O_2}$$
$$\mathrm{2(Cl + O_3 \longrightarrow ClO + O_2)}$$

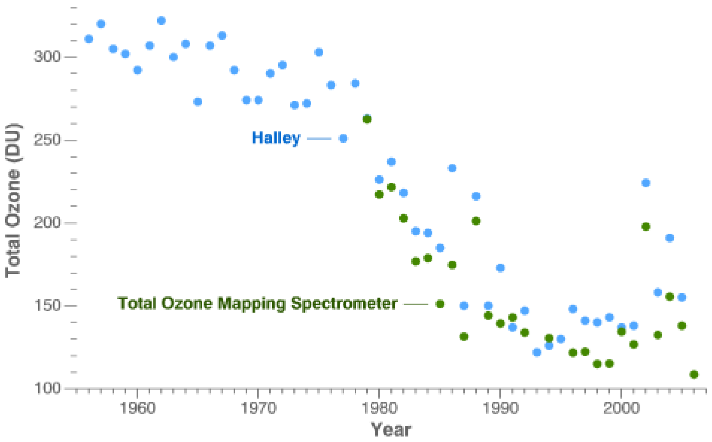
$$\text{NET: } \mathrm{2O_3 \longrightarrow 3O_2}$$

CYCLE 3 :

$$\mathrm{ClO + BrO \longrightarrow Cl + Br + O_2}$$
or
$$\left(\begin{array}{l}\mathrm{ClO + BrO \longrightarrow BrCl + O_2} \\ \mathrm{BrCl} + \textit{sunlight} \longrightarrow \mathrm{Cl + Br}\end{array}\right)$$
$$\mathrm{Cl + O_3 \longrightarrow ClO + O_2}$$
$$\mathrm{Br + O_3 \longrightarrow BrO + O_2}$$

$$\text{NET: } \mathrm{2O_3 \longrightarrow 3O_2}$$



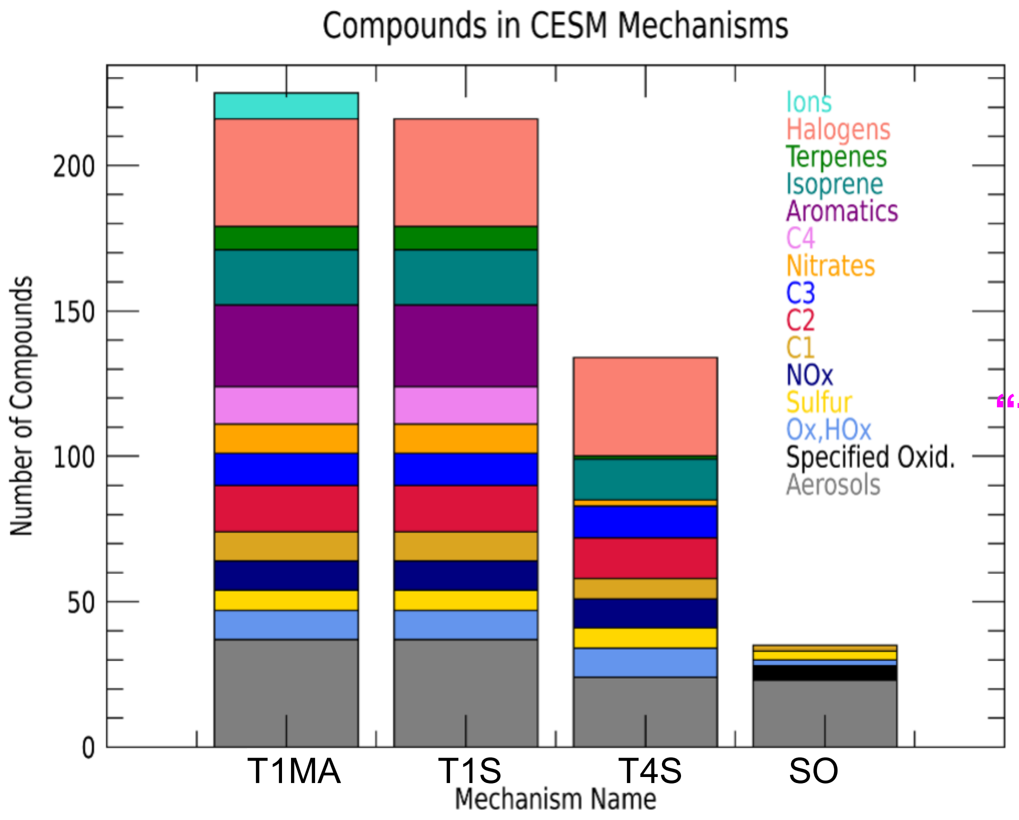


Comprehensive Stratospheric Chemistry

- Heterogeneous reactions
- Catalytic Cycles

Atmospheric chemistry mechanisms in CESM

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



T1S = default “full-chemistry” Troposphere and Stratosphere

Name	Description	# species
T1	Comprehensive tropospheric chemistry; for air quality simulations	179
T2	T1 with detailed terpene chemistry	265
T4	Simpler tropospheric chemistry suitable for climate simulations	97
T1S	T1 with comprehensive stratospheric chemistry	216
T1MA (TSMLT)	T1 with stratosphere, mesosphere, lower thermosphere chemistry	225
T4S	T4 with comprehensive stratospheric chemistry	134
SO	Specified Oxidants, with GHGs	33

Slide: Louisa Emmons

CAM6 (Specified Oxidants, SO) vs CAM -chem

Same atmosphere, physics, resolution

Different chemistry and aerosols -> emissions and coupling

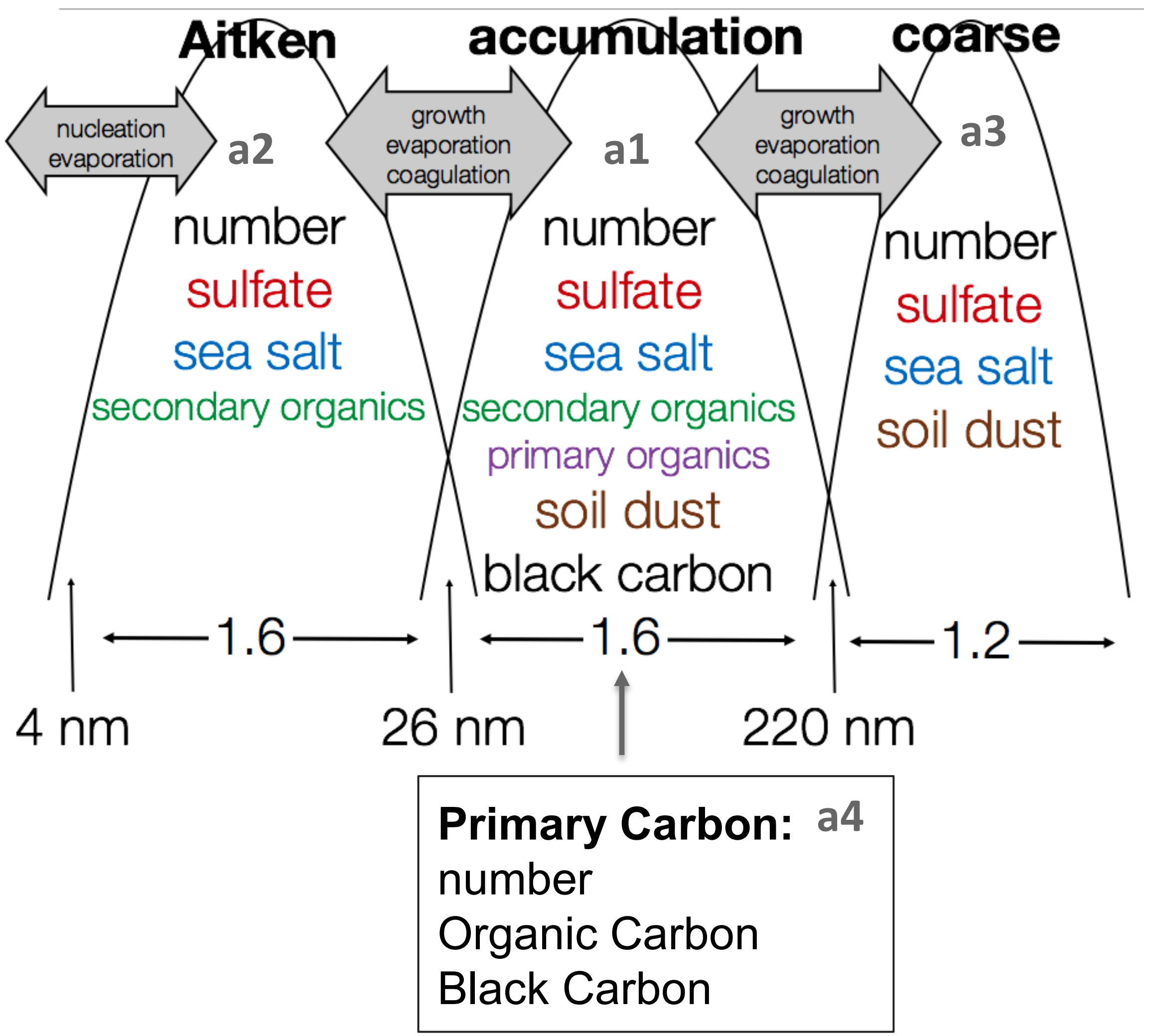
- **CAM6:** Aerosols are calculated, using simple chemistry (with "fixed" oxidants) (prescribed: N_2, O_2, H_2O, O_3, OH, NO_3, HO_2; chemically active: H_2O_2, H_2SO_4, SO_2, DMS, SOAG)

  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

Default Modal Aerosol Model (MAM4)

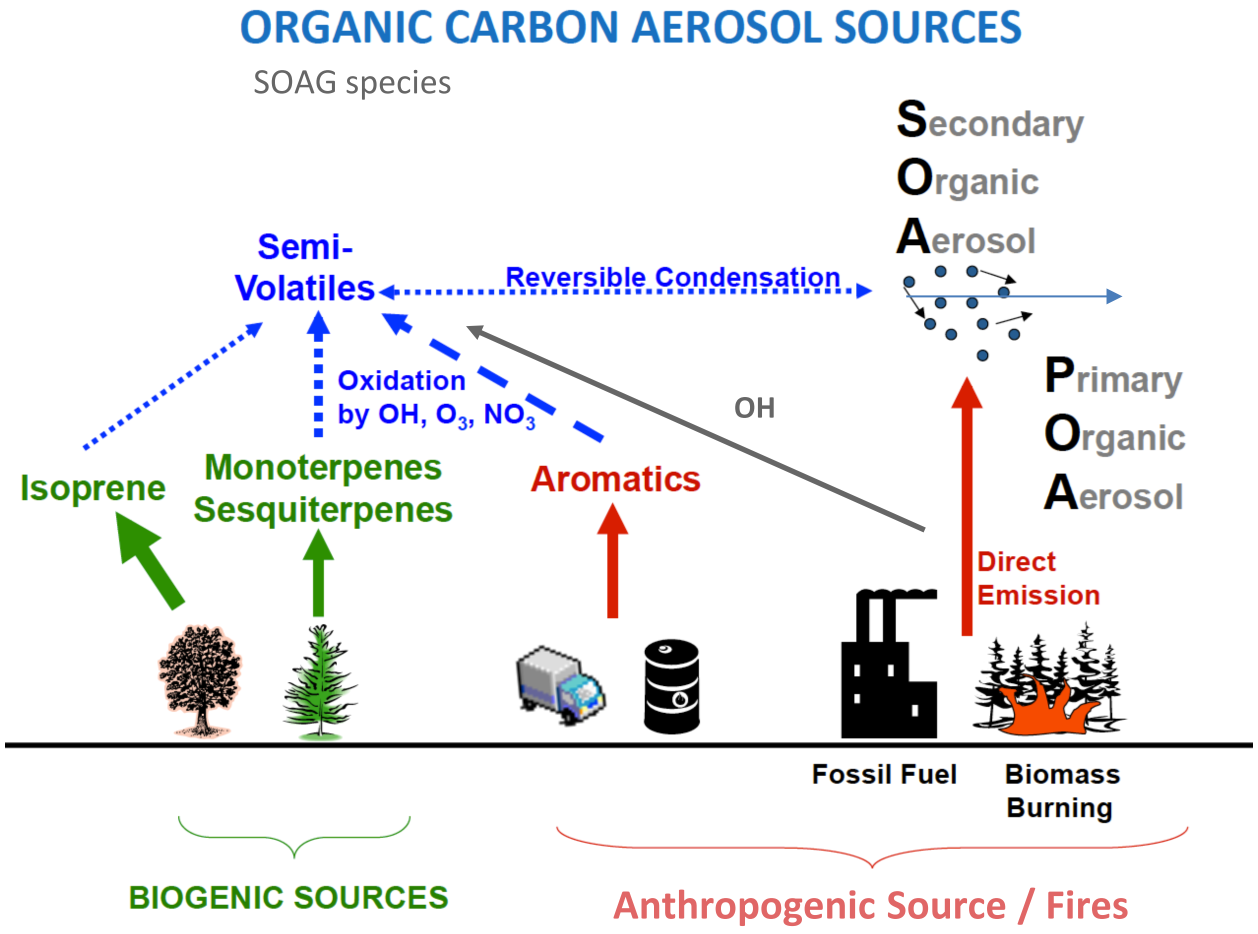


Representation of
- Sulfates,
- Black Carbon
- Organic Carbon, Organic Matter (OC, SOA),
- Mineral Dust and Sea-Salt

Slide: Mike Mills

Liu et al., 2016

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

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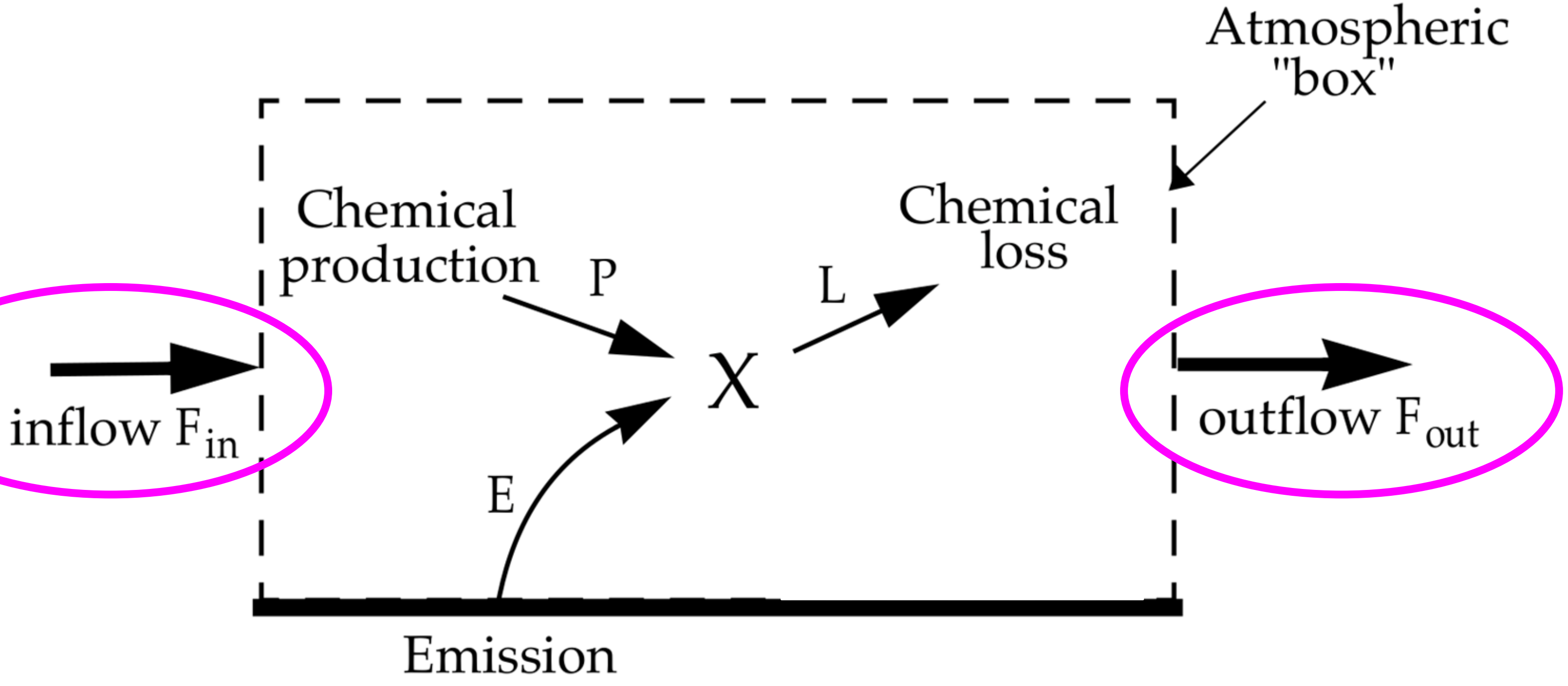
E_i Emissions

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 reminder

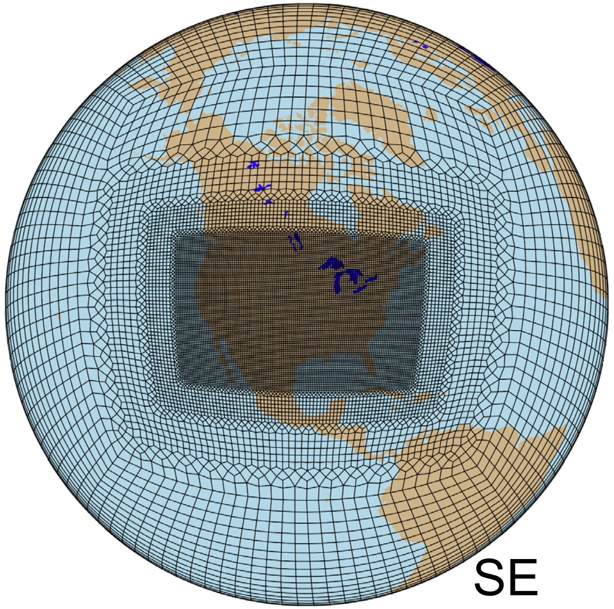
FV: Finite Volume (FV) “regular grid”

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

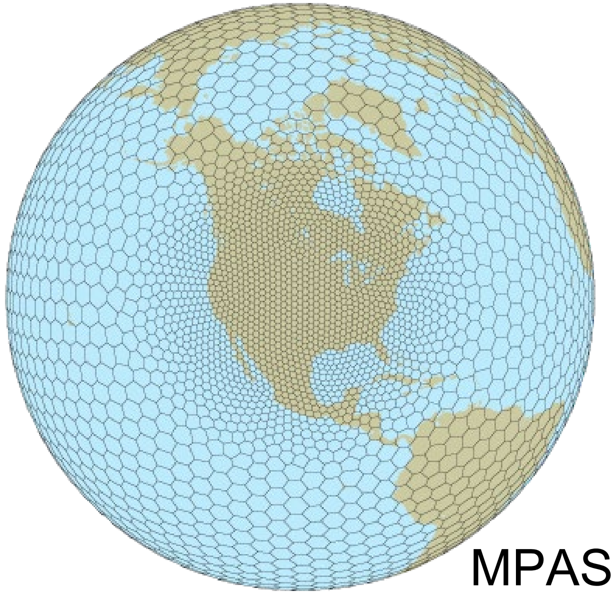
SE - CSLAM (pg3): Spectral Element dynamical core on a cubed sphere, Conservative Semi-Lagrangian Multi-tracer dynamical core with finite-volume transport (CSLAM). No current regional refined capability.

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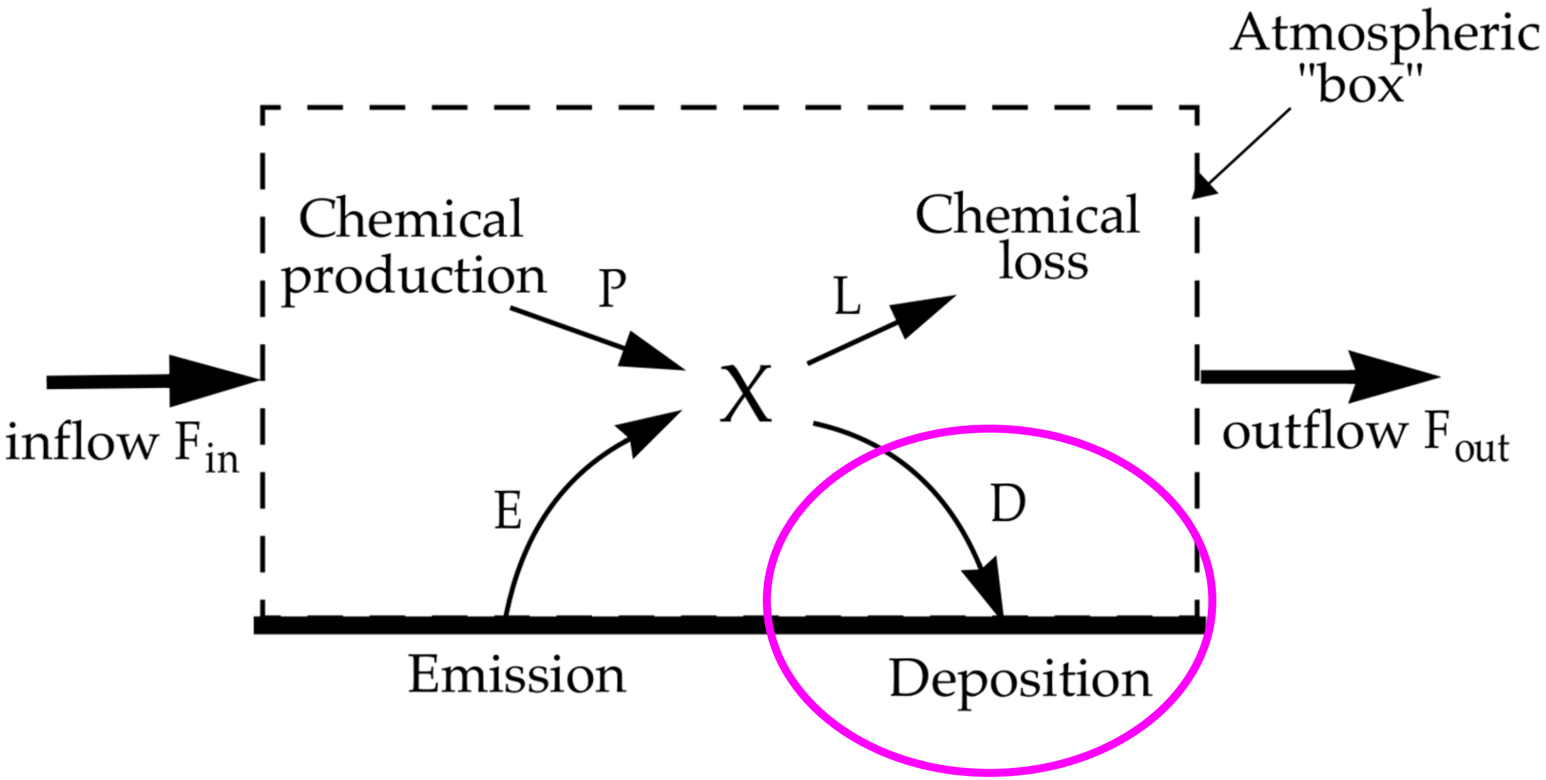
- E_i Emissions
- C_i Gas-phase-Chemistry
- A_i Aerosol-processes (Gas-aerosol exchange, het chem.)
- T_i Advection + Diffusion
- W_i Cloud-processes (wet deposition)
- D_i Dry deposition



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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

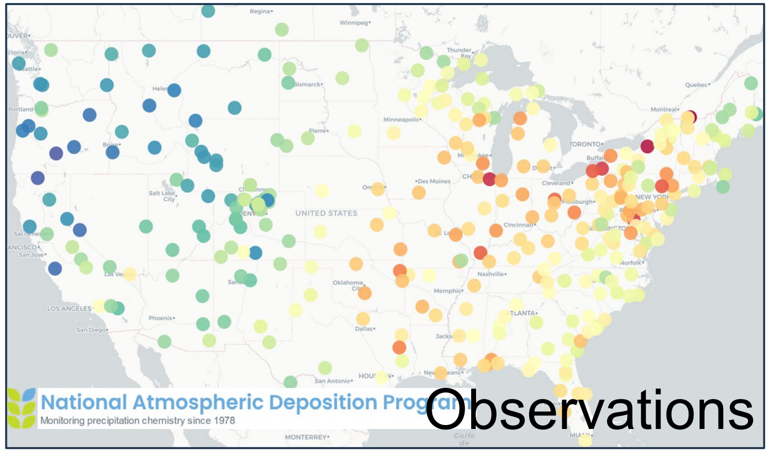
$$\chi_{iscav} = \chi_i \times F \times (1 - \exp(-\lambda \Delta t))$$

X_{iscav} scavenged species (kg)

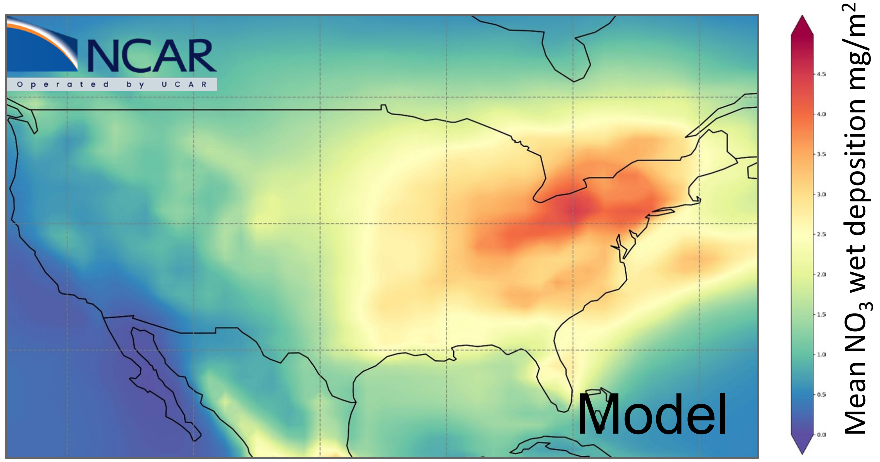
X_i species

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

λ is the loss rate

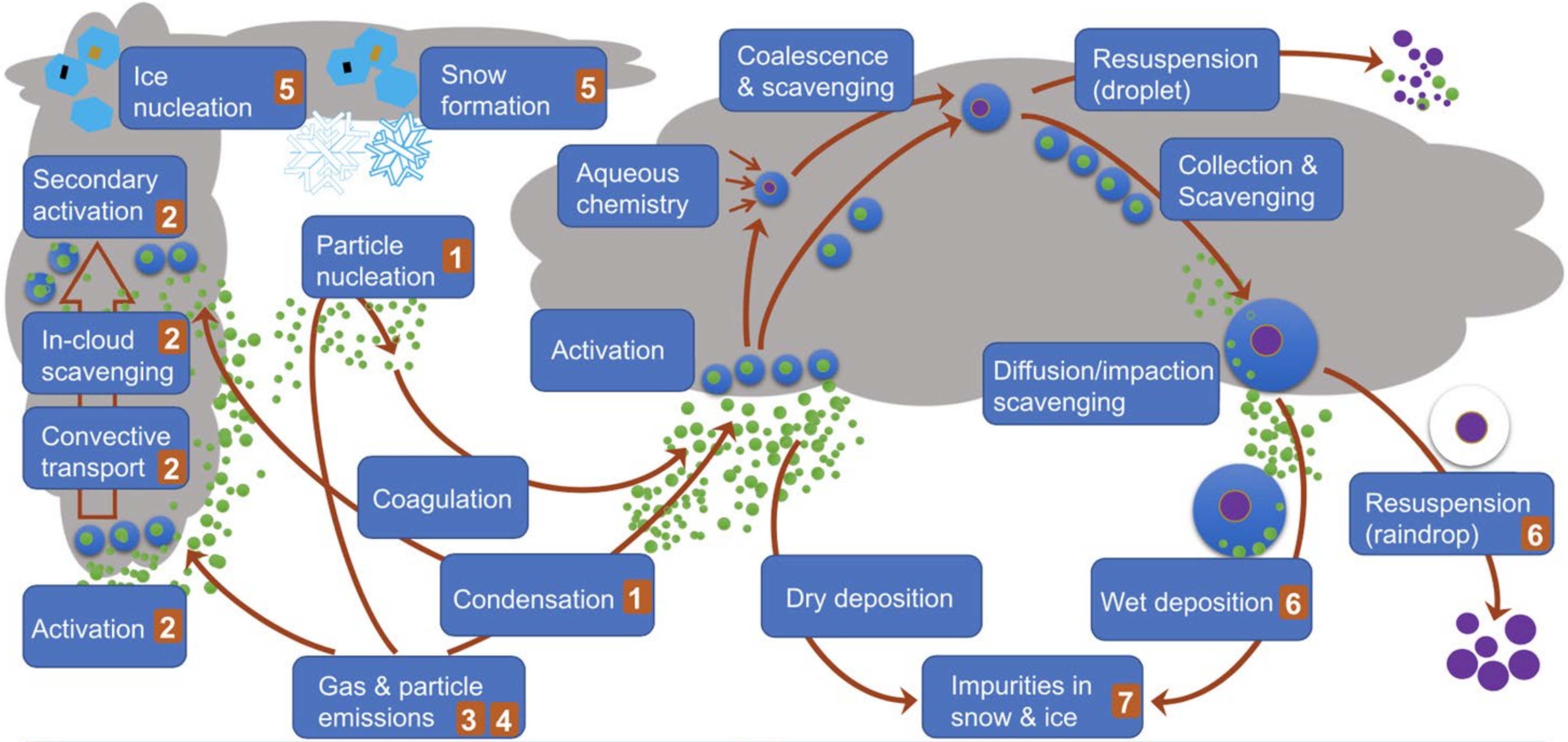


Deni Murray ACOM ASP graduate visitor



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

Aerosol – Cloud Interactions



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

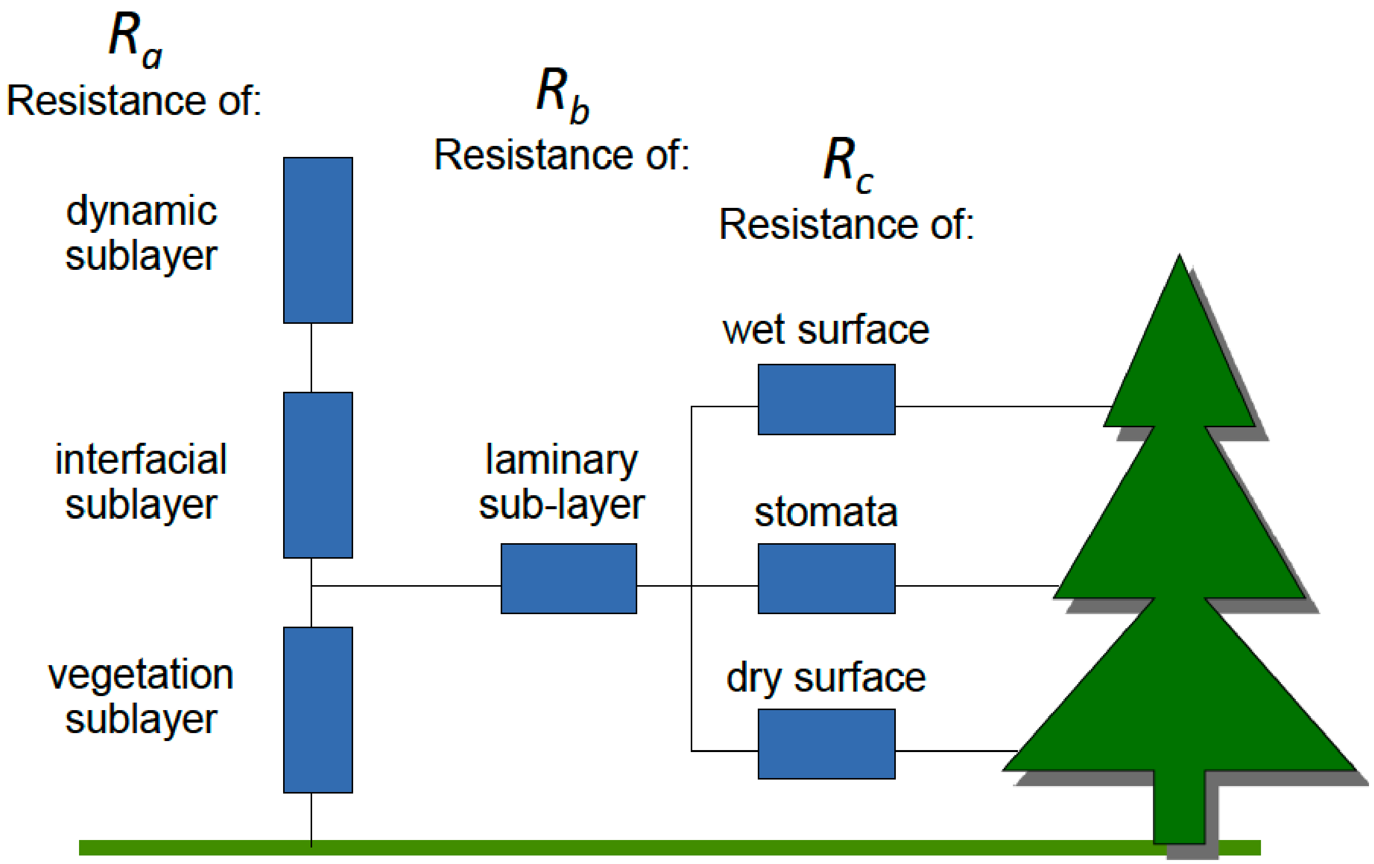
Resistance model:

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

$$F = -v_d C$$

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



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it can get expensive very fast! $$$

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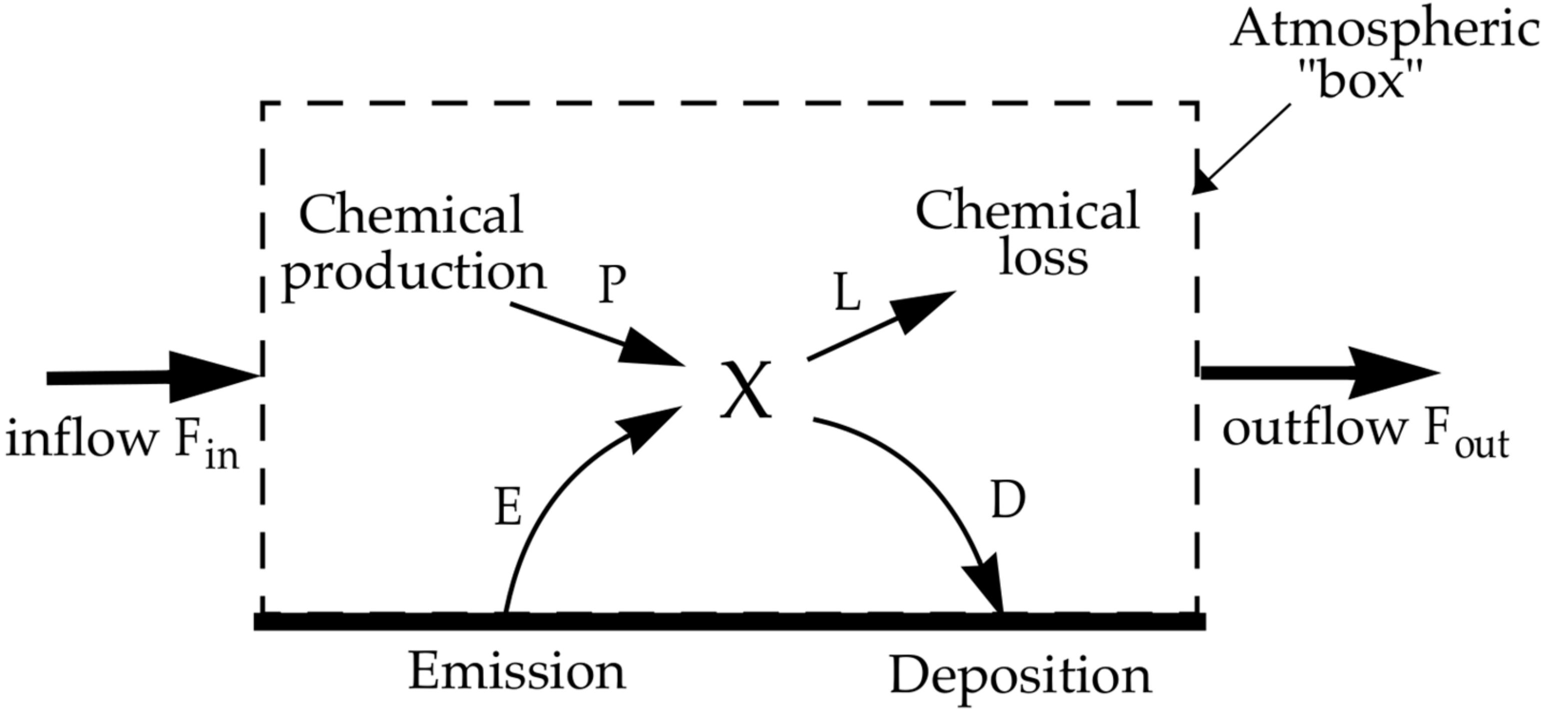
- A_i Aerosol-processes (Gas-aerosol exchange, het chem.)
- T_i Advection + Diffusion
- W_i Cloud-processes (wet deposition)
- D_i Dry deposition



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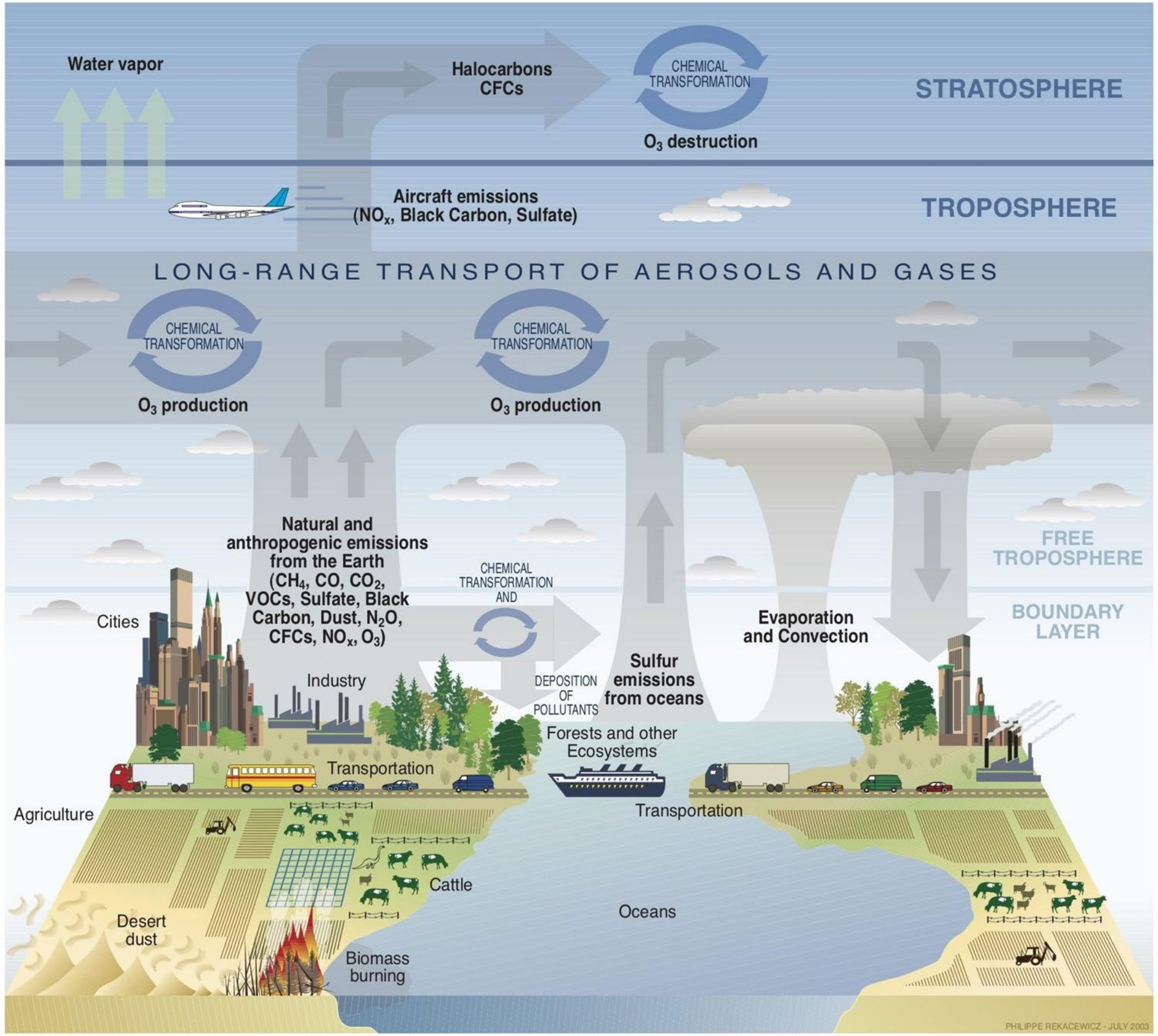
- Applications: CAM-chem
- Support

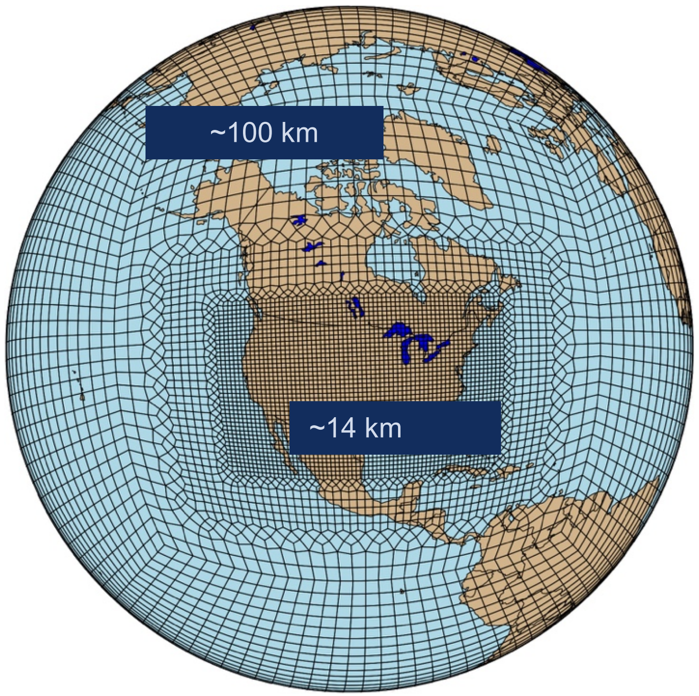


Chemistry → Air Quality: Regional refinement

MUSICA-V0: **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

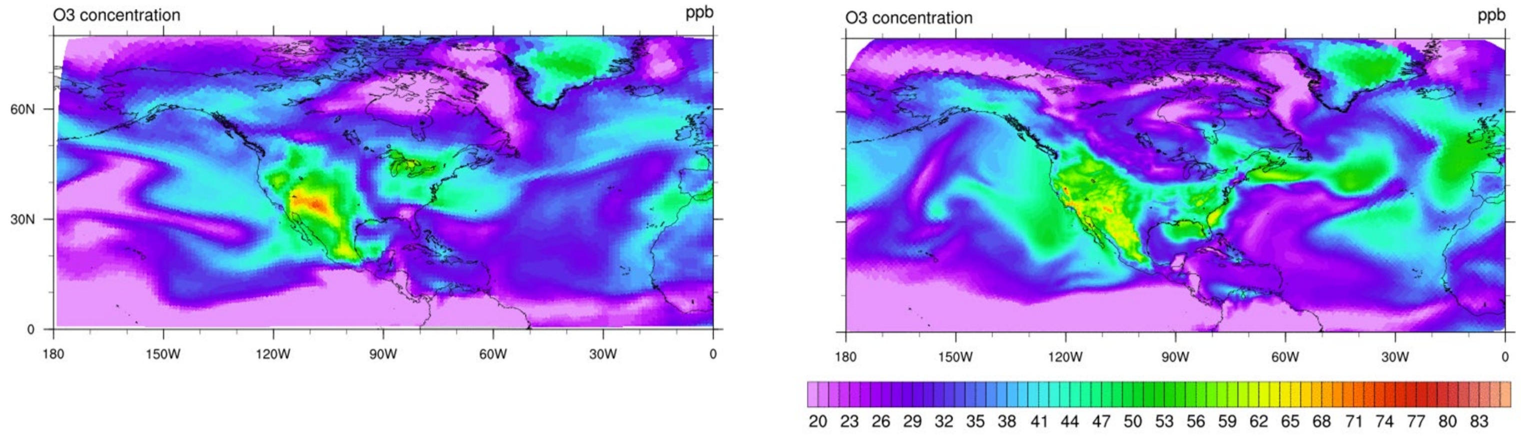


Example: U.S. Air Quality, Surface Ozone (ppb)

- Exposure Relevant scales and large-scale feedbacks

Global 1 degree

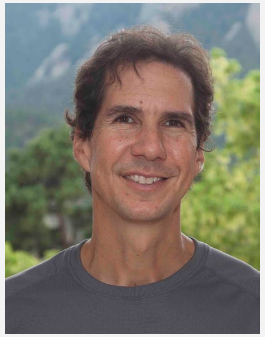
Regional Refined



Credits: Forrest Lacey (ACOM)

Chemistry → Climate: Australian wildfires 2019/2020

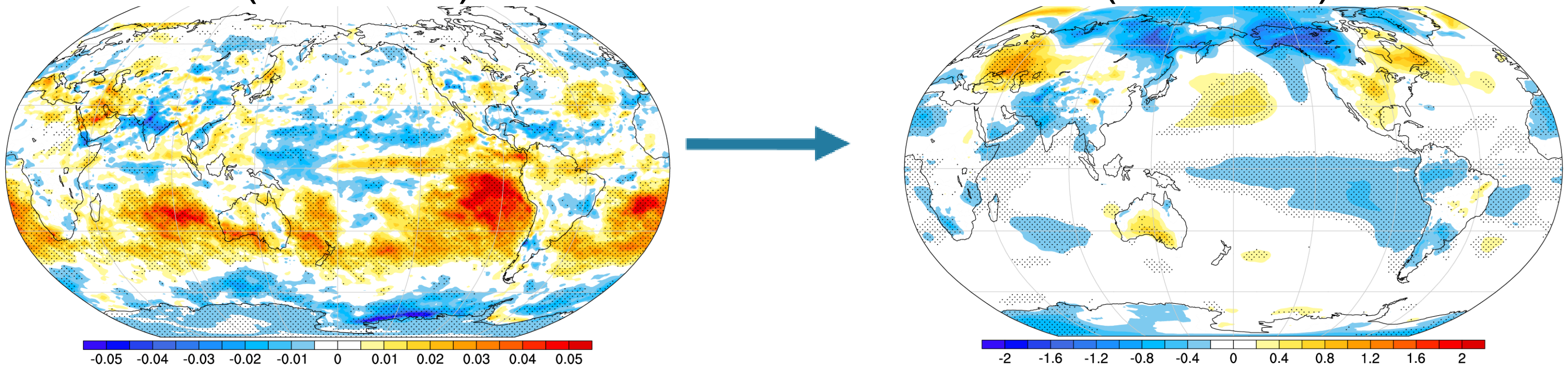
- CESM/CAM6 simulation with aerosols, satellite-based inventory (GFED) in Australia compared to climatology
- Climate response similar to a major volcanic eruption (aerosol-cloud interactions)
- Large interhemispheric radiative imbalance anomaly and impacts on ENSO

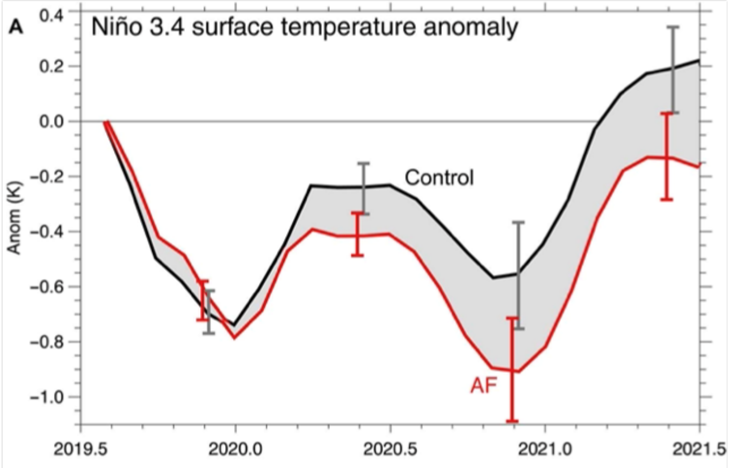


John Fasullo, CGD

Cloudy Sky Albedo (Jan 2020)

Near-surface temperature (Jan 2021)





Cloud brightening across the Southern Hemisphere

2020/21 La Niña response

Fasullo et al., GRL, 2021

Fasullo et al., Sci. Adv., 2023

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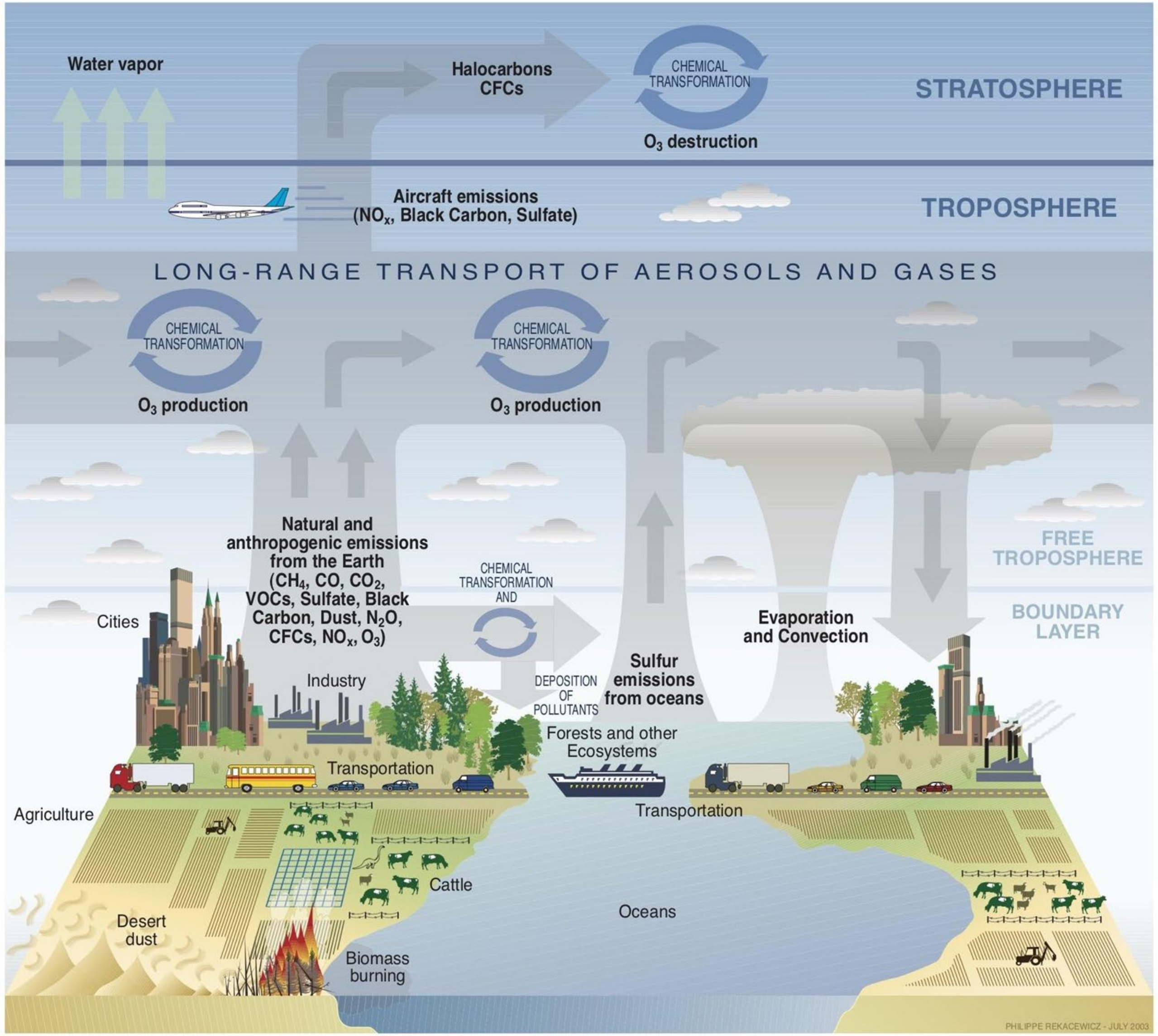

User Support for chemistry modeling in CESM

Wiki Page for Chemistry:
https://wiki.ucar.edu/display/camchem/Home

Use and Diagnostics	• Boundary conditions for regional modeling • Atmospheric Diagnostics (ADF) in python ***NEW!*** • Automated CESM diagnostic package (using NCL) • Using CAM-chem output • MELODIES MONET model-obs comparison package
User Community	• Current Users/Projects • Contributions to Model Intercomparisons (MIPs) • CAM-chem Forum • Chemistry-Climate Working Group Publications • CAM-chem Publications from NCAR • CESM Publications
Other links and documents	• Recent Bug Fixes • CAM Documentation (User and Scientific Guides) • ACOM CAM-chem page • CESM Chemistry Climate Working Group • Join the CESM Chemistry WG mailing list • Benchmarks and Production Experiment Diagnostics

Regional Refinement WIki: https://wiki.ucar.edu/display/MUSICA

Forum to search for and ask questions:
http://bb.cgd.ucar.edu /

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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**

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

  - 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 and model configuration is important to correctly answer your question.