Modeling Tropospheric Chemistry: 0D to 3D models

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Fundamentals of Atmospheric Chemistry and Aerosol Modeling

Aug 13-15, 2018

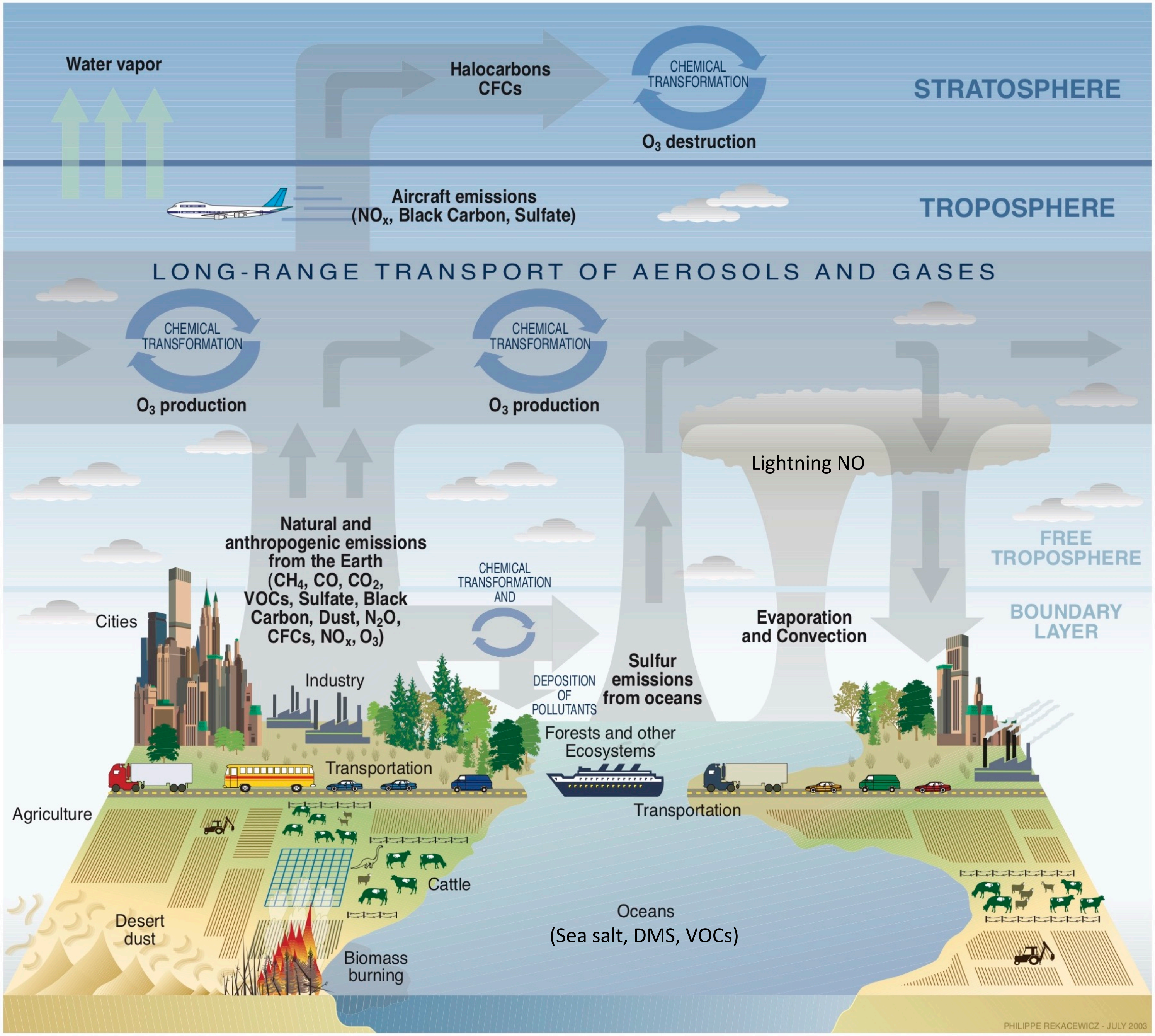


Fig. 3-1 of Climate Change Science Program Strategic Plan, 2003

Some regions, e.g. the Arctic, require unique considerations …

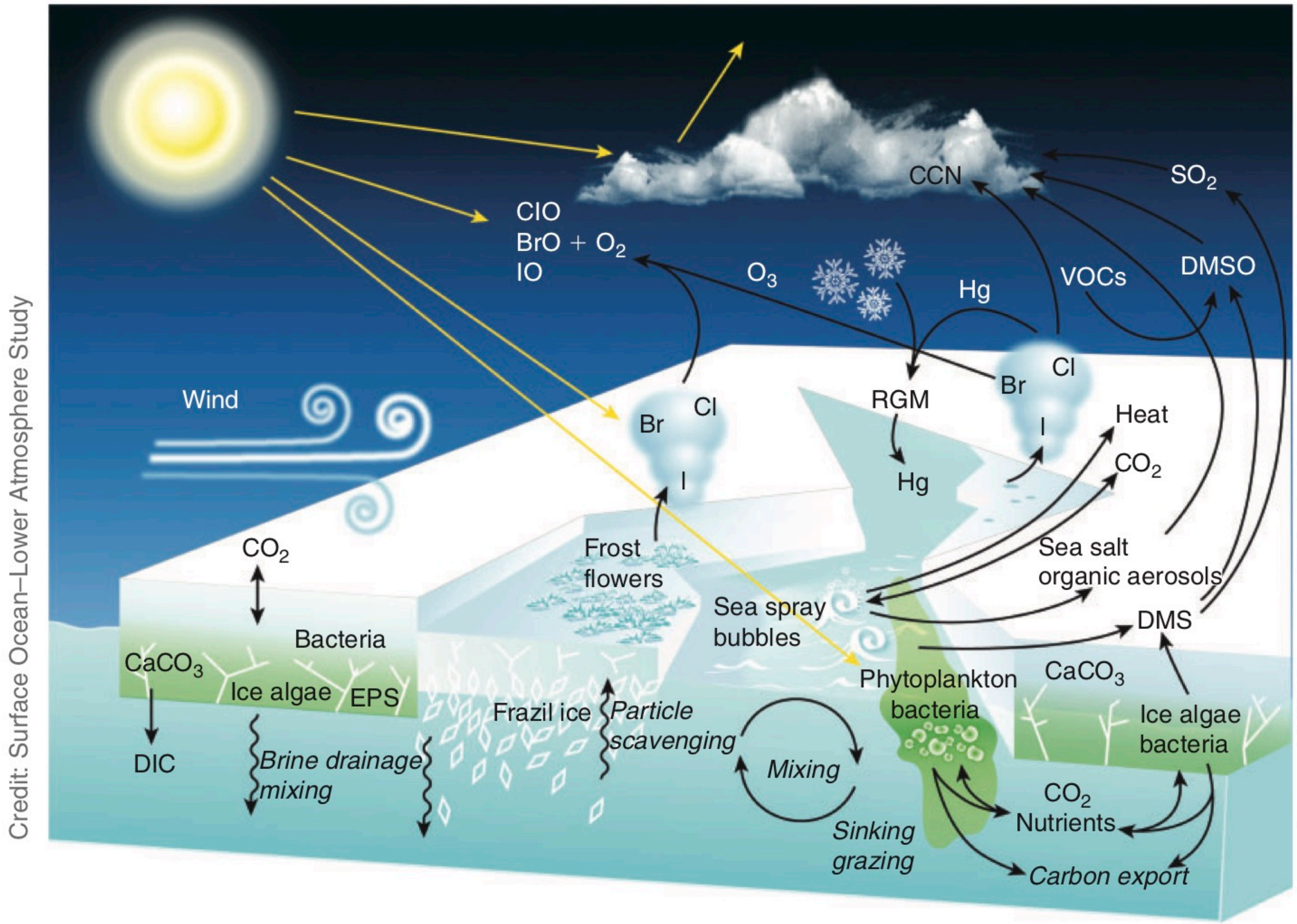
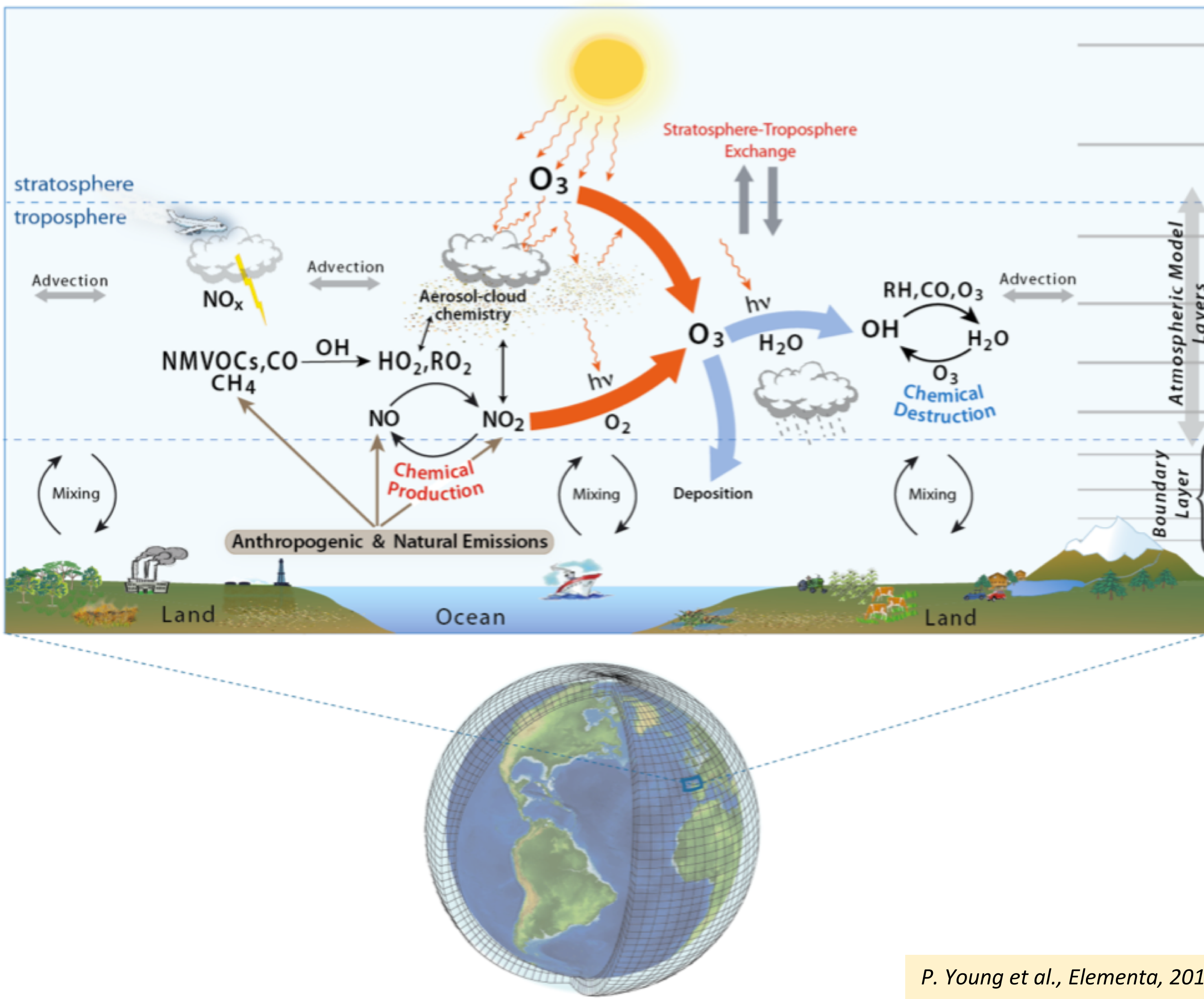


Fig. 3. Conceptual diagram illustrating the main issues, processes and species relating to the SOLAS Sea Ice strategy (after Shepson et al.[54]). Processes are indicated in italic. (DIC, dissolved inorganic carbon; EPS, extracellular polymeric substances; CCN, cloud condensation nuclei; RGM, reactive gaseous mercury; VOCs, volatile organic compounds; DMS, dimethyl sulfide; DMSO, dimethyl sulfoxide.)

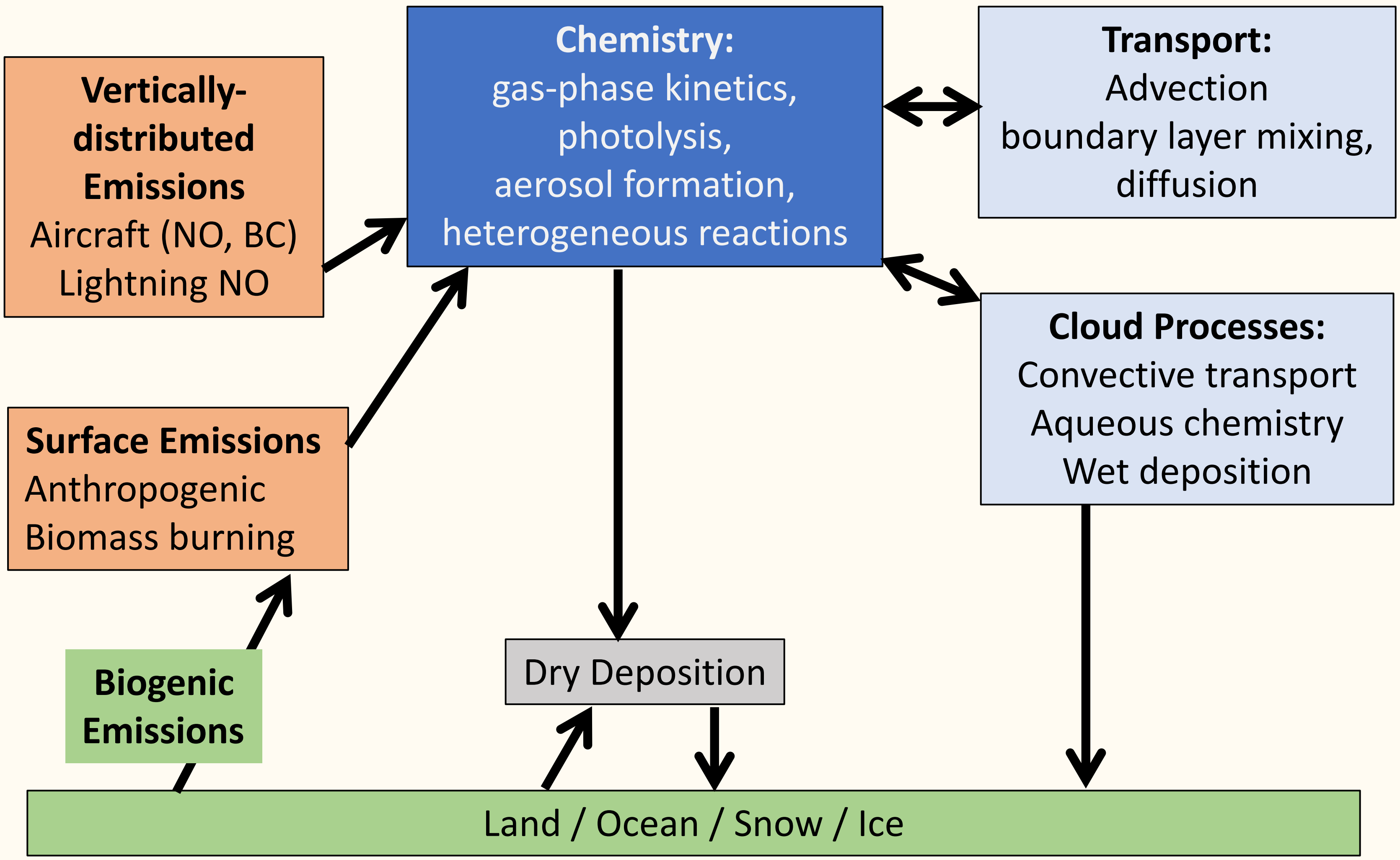
C.S. Law et al., Environ. Chem., 2013

Simulating tropospheric ozone



P. Young et al., Elementa, 2017

Computer models treat each process as a separate module



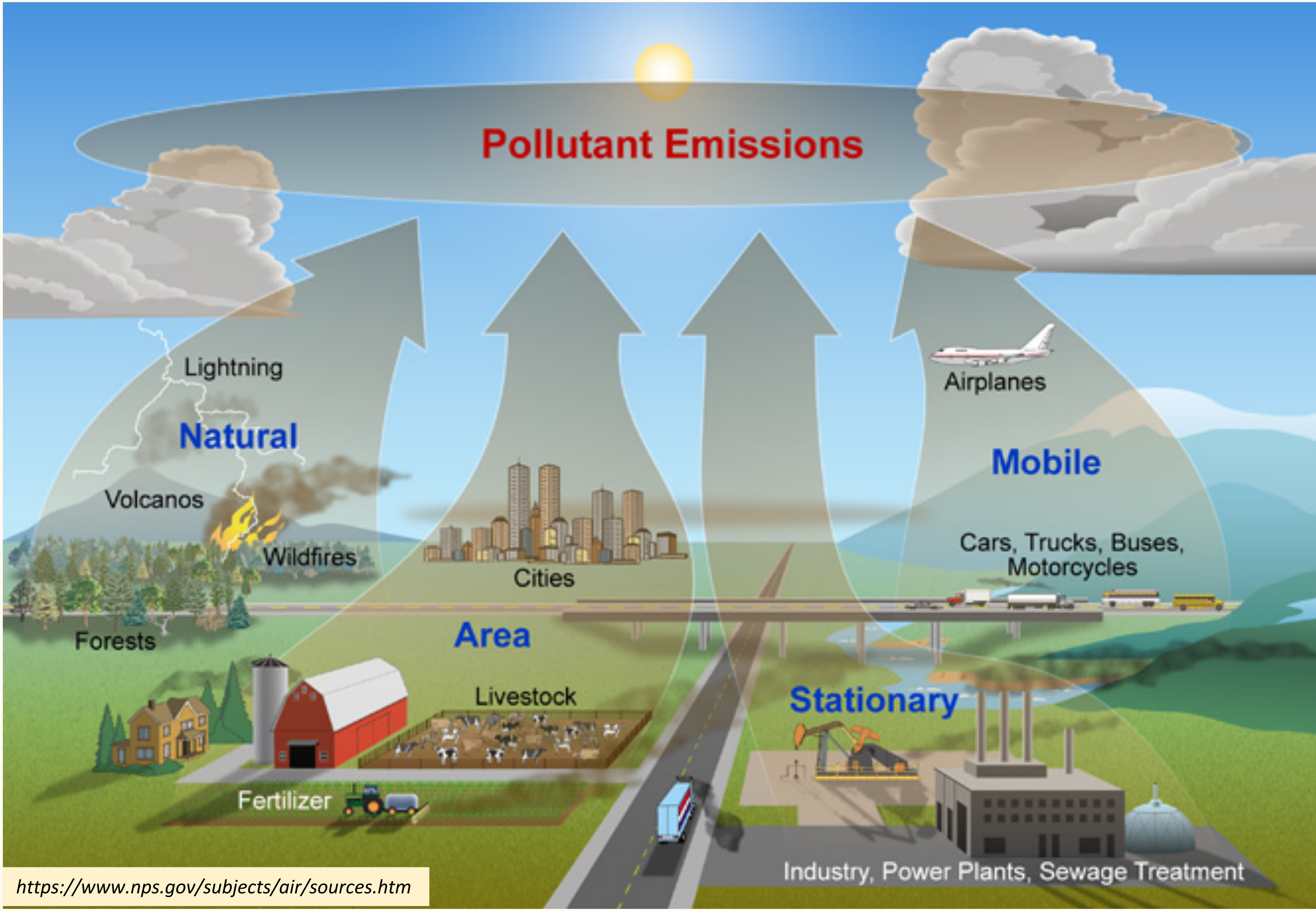
Solution for each chemical species *i*

$$\frac{\partial c(i)}{\partial t} = Production(i) - Loss(i) = E_i + C_i + A_i + T_i + W_i + D_i$$

For each compound, at each timestep, the change in concentration is the sum of the change in concentration for each process:

E_i: Emissions
C_i: Gas-phase-Chemistry
A_i: Aerosol-processes
T_i: Advection + Diffusion
W_i: Cloud-processes (wet deposition)
D_i: Dry deposition

For compounds with short lifetimes the order of operators can affect results


Pollutant Emissions
Lightning
Natural
Volcanos
Wildfires
Forests
Cities
Area
Livestock
Fertilizer
Airplanes
Mobile
Cars, Trucks, Buses, Motorcycles
Stationary
Industry, Power Plants, Sewage Treatment
https://www.nps.gov/subjects/air/sources.htm

Anthropogenic emissions

- Power plants (NO_x, SO_2)
- Industry (solvents – VOCs)
- Traffic (NO_x, CO, PM)
- Residential heating (CO, VOCs, PM)
- Agriculture (NH_3)
- Shipping (NO_x, HCs)
- Aviation (NO_x)

Many emissions inventories at: http://eccad.aeris-data.fr/

ECCAD: Emissions of atmospheric Compounds and Compilation of Ancillary Data

En Fr

Sign in

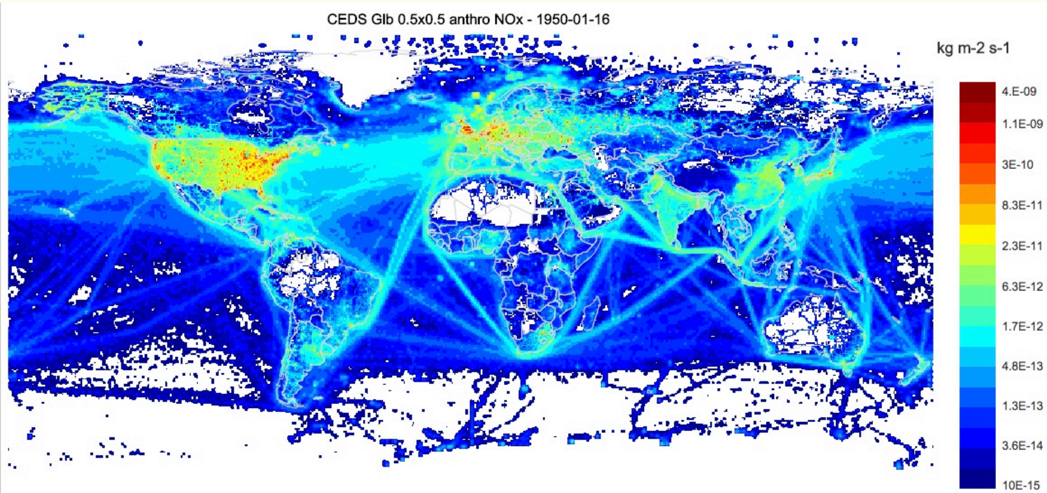
Title	Categories	Temporal coverage	Time resolution	Grid size	Provider(s)
MACCity Global					
RCPs Global					
ACCMIP Global					
CAMS-GLOB-ANT Global					
CAMS-GLOB-BIO Global					
CAMS-GLOB-SHIP Global -Pole					
CAMS-GLOB-OCE Global					
CAMS-REG-GHG TNO Europe					
CAMS-REG-AP TNO Europe					
HTAPv2 Global					
EDGARv4.3.2 Global					
EDGARv4.3.1 Global					
EDGARv4.2 Global					
EDGARv4.3.2-monthly Global					
EDGARv4.3.1-monthly Global					
ECLIPSE-GAINS-4a Global					
ECLIPSE-GAINS-V5a Global					
CEDS Global					
RETRO Global					
Junker-Liousse Global					
Andres-CO2-v2016 Global					

Title	Categories	Temporal coverage	Time resolution	Grid size	Provider(s)
HYDE1.3 Global	Anthropogenic	1890-1990	Monthly	1°	EDGAR
POET Global	Anthropogenic Biomass burning Biogenic Oceanic	1990-2000 1990-2000 1990-2000 1990-2000	Yearly Monthly Monthly Yearly	1°	
GEIA Global	Anthropogenic Biomass burning Biogenic Oceanic Volcanic Lightning	1984-2018 1984-1990 2000-2000 1990-2000 2000-2000 1990-1990	Yearly Yearly Yearly Yearly Yearly Monthly	1°	GEIA
EDGARv4.tox1 Global	Anthropogenic	1970-2008	Yearly	0.1°	EDGAR
GFASv1.3 Global	Biomass burning	2003-2016	Daily	0.1°	macc
GFASv1.2 Global	Biomass burning	2003-2015	Daily	0.1°	macc
GFED3 Global	Biomass burning	1997-2010	Monthly	0.5°	GFED
GFED4 Global	Biomass burning	1997-2015	Monthly	0.25°	GFED
IS4FIRES Global	Biomass burning	2000-2011	Monthly	0.5°	
DECSO-NOx China South Africa Middle East					
PKU05 Global					
PKU Global					
GICC Global					
GUESS-ES Global					
AMMABB Global					
MEGAN-MACC Global					
MEGANv2 Global					
APIFLAME Charmex					
TNO-MACCII TNO Europe					

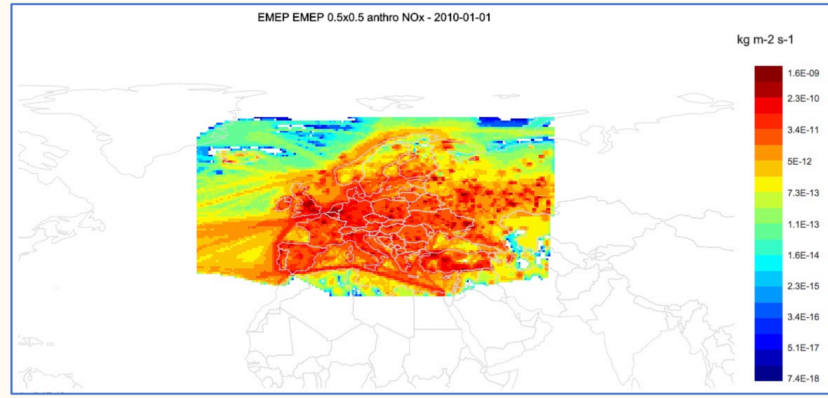
Title	Categories	Temporal coverage	Time resolution	Grid size	Provider(s)
EMEP EMEP Europe	Anthropogenic	1980-2020	Yearly	0.5°	emep
REAS1.1 East Asia	Anthropogenic	1980-2020	Monthly	0.5°	
MPI-CNRS East Asia 2	Anthropogenic	2008-2016	Yearly	0.25°	
CR2-MMA Chile	Anthropogenic	2016-2016	Yearly	0.01°	(CR)²
REAS2.1 East Asia	Anthropogenic	2000-2008	Monthly	0.25°	
IASB-TD-OMI Global	Biomass burning Biogenic	2005-2014 2005-2014	Monthly Monthly	0.5°	
IASB-TD-OMI-NCP North China Plain	Anthropogenic	2007-2012	Monthly	0.25°	
MarcoPoloKNMI China	Anthropogenic	2007-2013	Yearly	0.25°	GLOBEMISSION
L14-Africa Africa	Anthropogenic	2005-2030	Decadal	0.25°	
SAFAR-India India	Anthropogenic	1991-2011	Monthly	1°	
DACCIWA Africa	Anthropogenic	1990-2015	Yearly	0.125°	
DACCIWA-flaring Africa	Anthropogenic	1994-2015	Yearly	0.125°	

Anthropogenic NO_x Emissions – various inventories

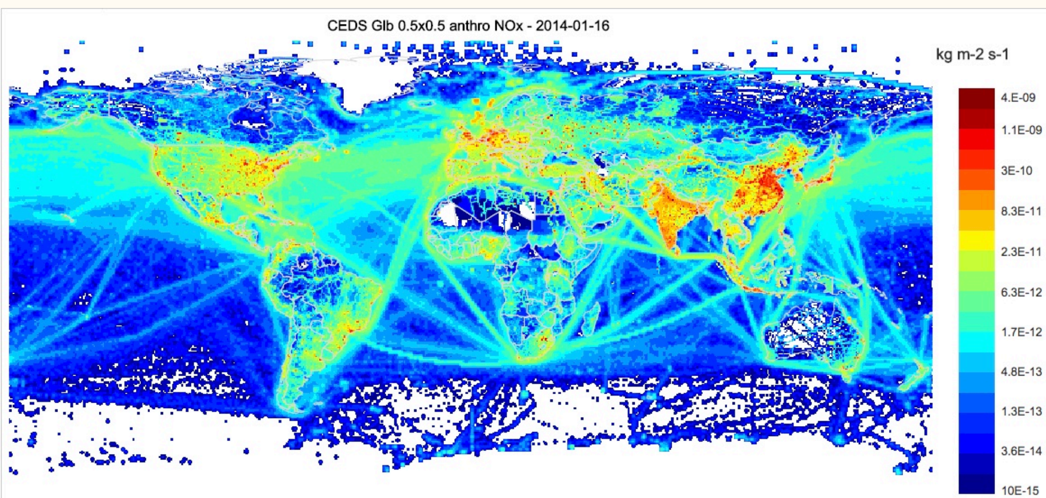
CEDS (0.5deg) - 1950



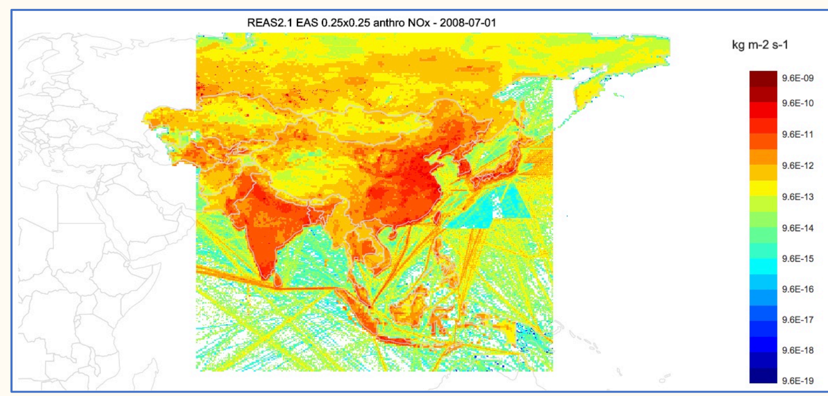
EMEP Europe (0.5 deg) - 2010



CEDS - 2014



REAS2.1 Asia (0.25deg) - 2008



Images from: http://eccad.aeris-data.fr/

Large differences between inventories

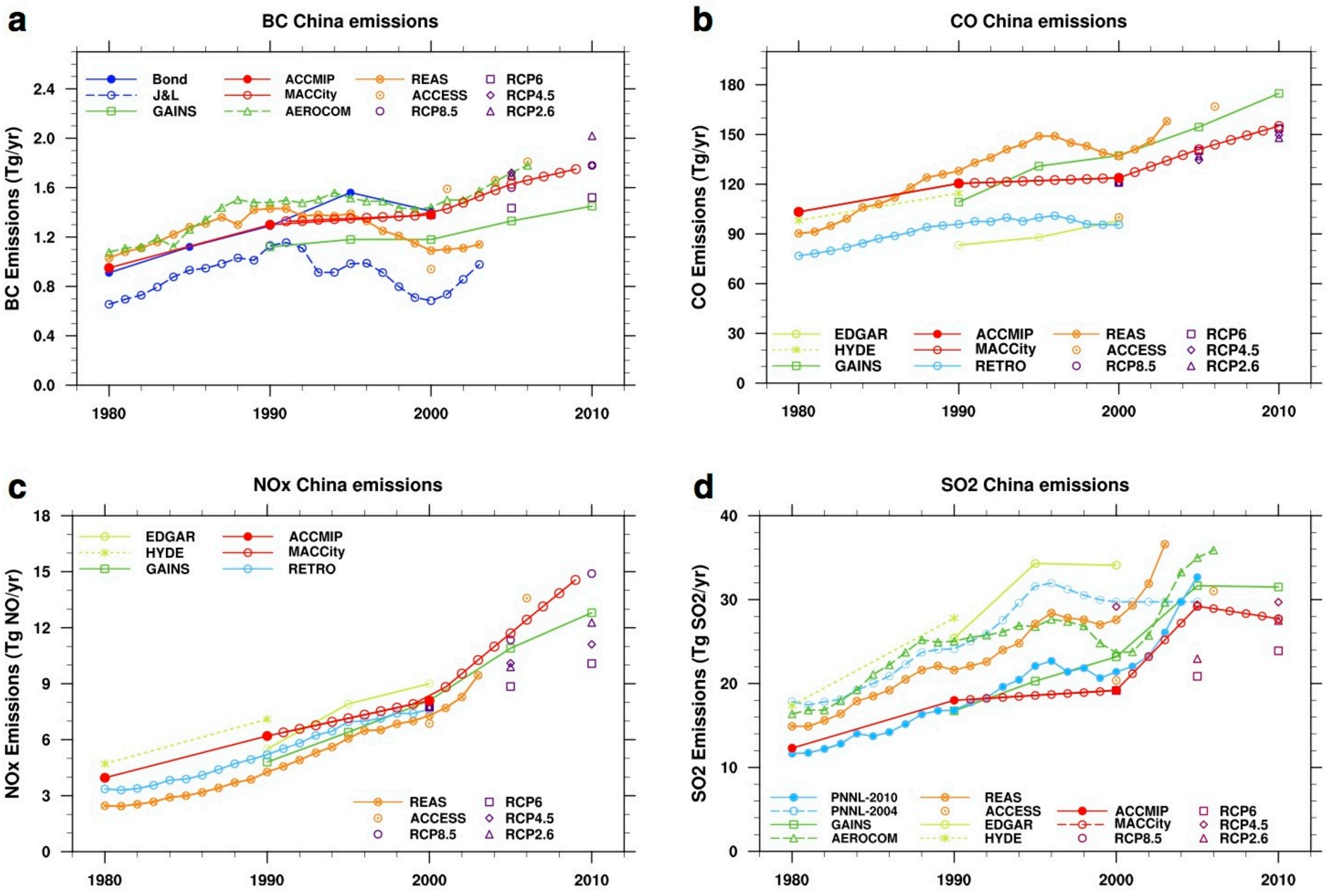


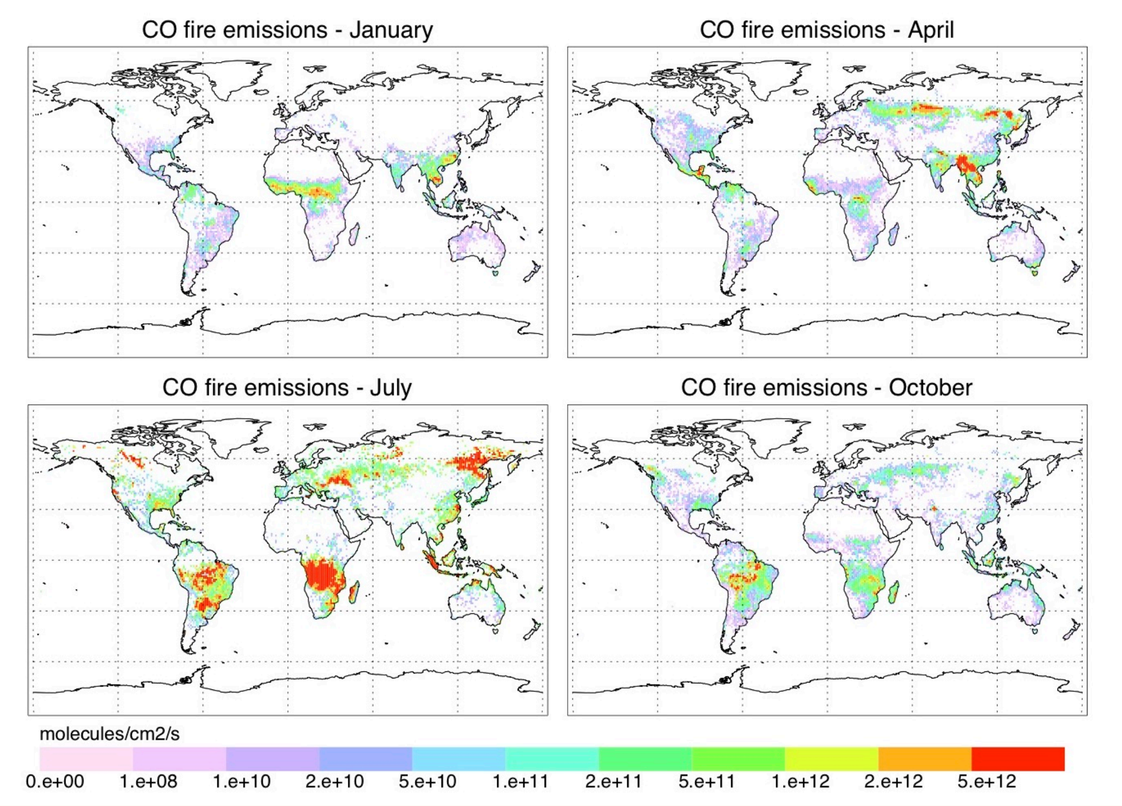
Fig. 4 Emissions of BC (*top left*), CO (*to right*), NO_x (*bottom left*) and SO_2 (*bottom right*) in China from 1980 to 2010

Granier et al., Climatic Change, 2011

Biomass burning

Natural and Anthropogenic

- Wildfires
- Prescribed burns (forest understory)
- Agricultural burning
- Trash burning
- Residential burning of biofuel



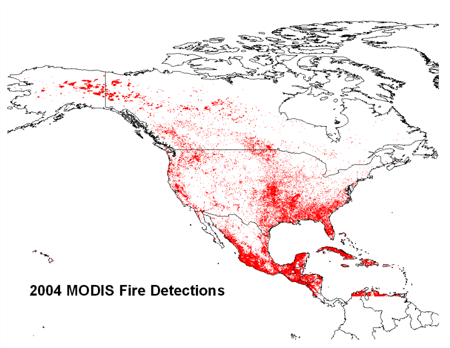
Large seasonal variability
- Natural wet/dry season
- Agricultural activity

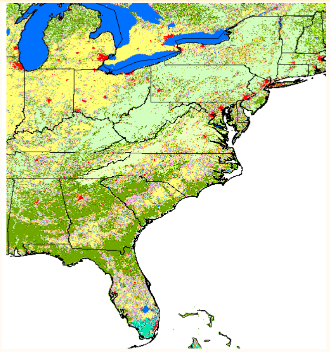
Large inter-annual variability due to climate

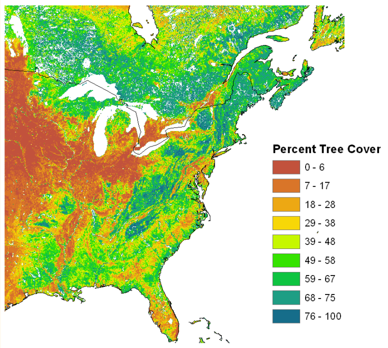
Large uncertainties in
- What is burned
- How much is burned
- Emission factors

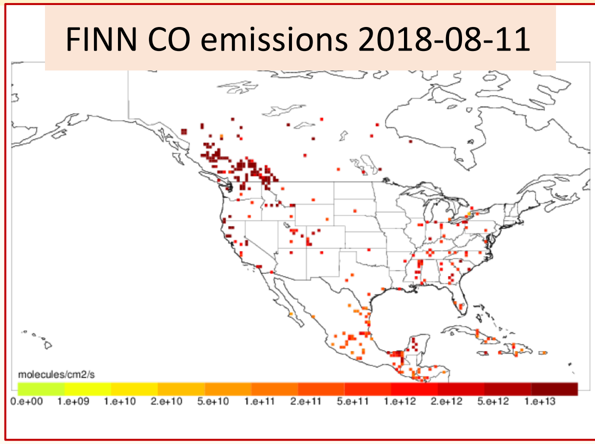
Wiedinmyer et al., GMD, 2011

Fire emissions models (e.g., FINN)









Emissions(i) = f(A(x,t), B(x,t), E_f(i))

For compound *i*, location *x*, time *t*:

- A(x,t): Area burned
- B(x,t): Biomass burned (kg-biomass-burned/area)
- E_f: Emission factor for each chemical compound (mass-emission-species/kg-biomass-burned)
  - type of vegetation (ecology)
  - fuel characteristics (amounts of each type of biomass)
  - fuel condition (moisture content)

Input data sets

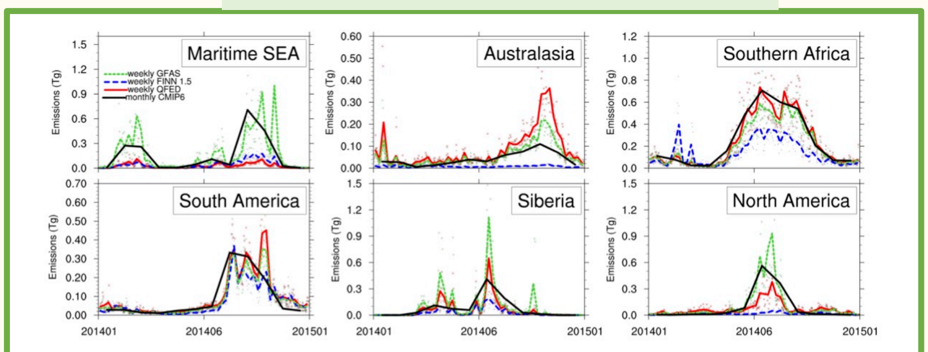
- MODIS Thermal Anomalies (Fire counts)
- Land cover maps (from MODIS and SPOT)

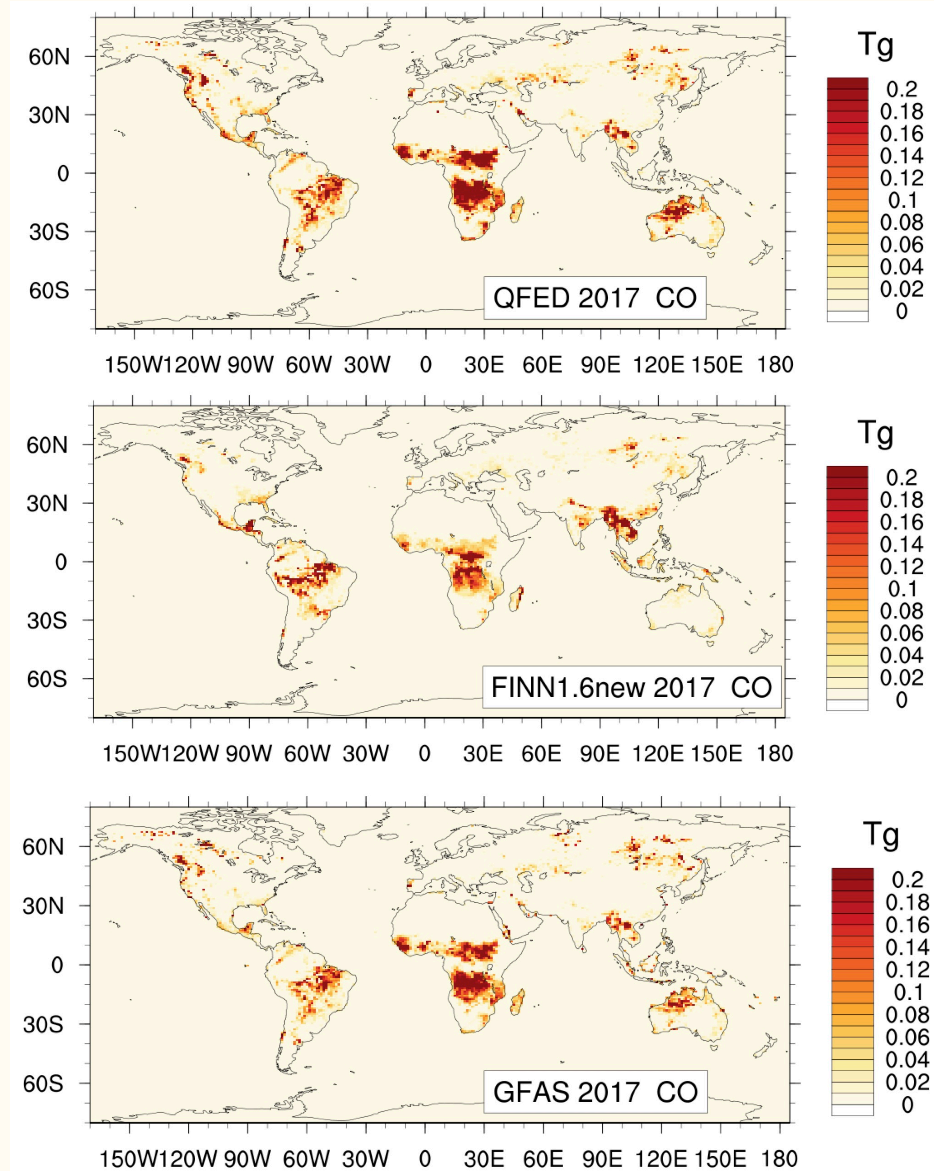
Other inventories use similar algorithms
Some use Fire Radiative Power satellite obs
Different landcover maps, emission factors, etc.

Differences in fire inventories

- FINN- Fire INventory from NCAR (Wiedinmyer)
- QFED - Quick Fire Emissions Dataset (NASA)
- GFAS – Global Fire Assimilation System (CAMS/ECMWF)
- GFED – Global Fire Emissions Database (van der Werf et al.)
- For CMIP6 (1750-2100)

Monthly fire emissions - 2014





Natural emissions

Natural terrestrial emissions

- VOCs from vegetation (isoprene, terpenes, methanol, etc.)
- Soil NO
- Volcanoes (SO_2)

Lightning NO

Ocean emissions – DMS, CO, VOCs

Dust

Sea salt

A biogenic emissions model: MEGANv2.1

Inputs: model-simulated data or observations

LAI: Leaf Area Index
PFT: Plant Functional Type

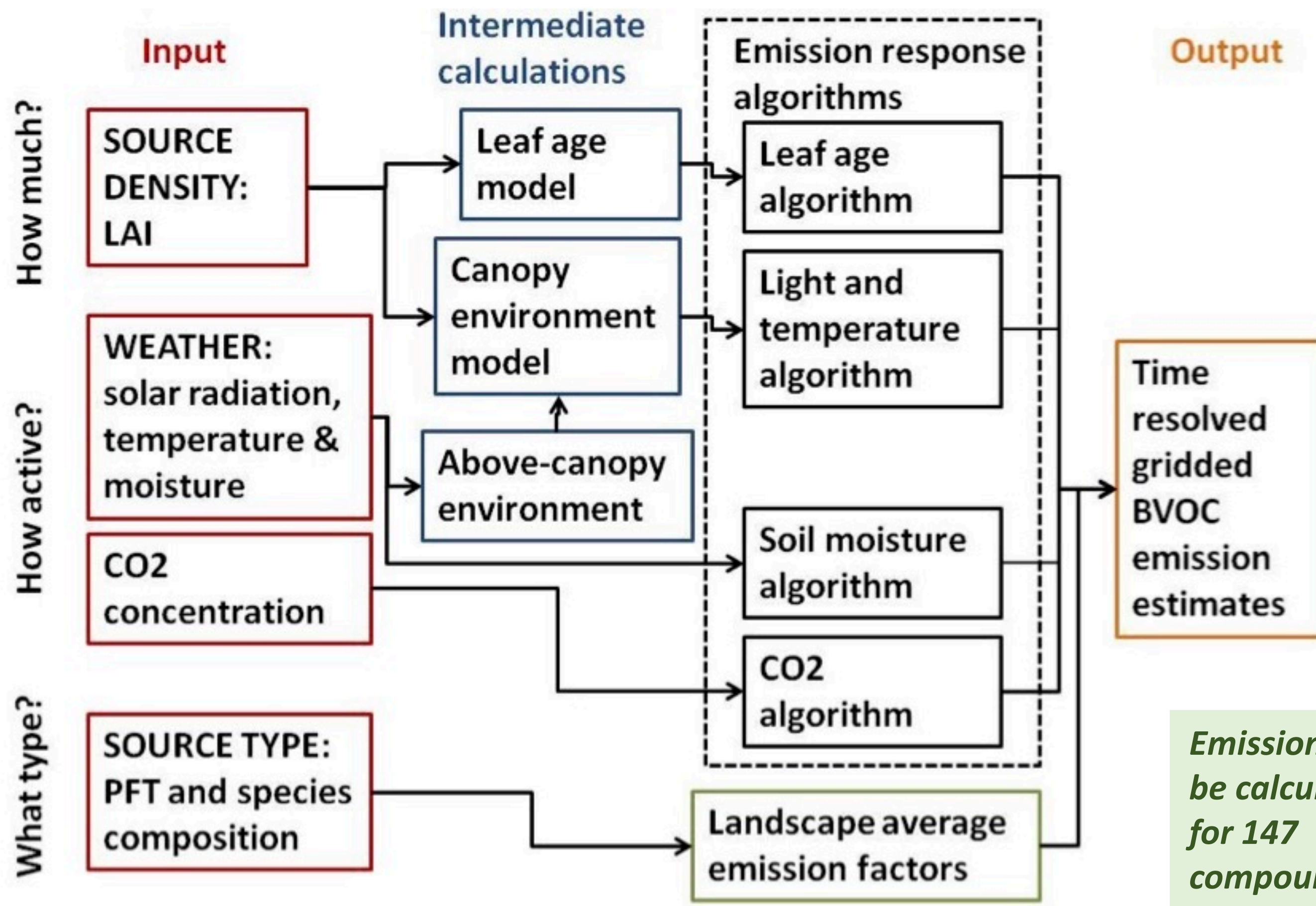


Fig. 1. Schematic of MEGAN2.1 model components and driving variables.

Emissions can be calculated for 147 compounds

Guenther et al., GMD, 2012

MEGAN algorithm in a 3D model

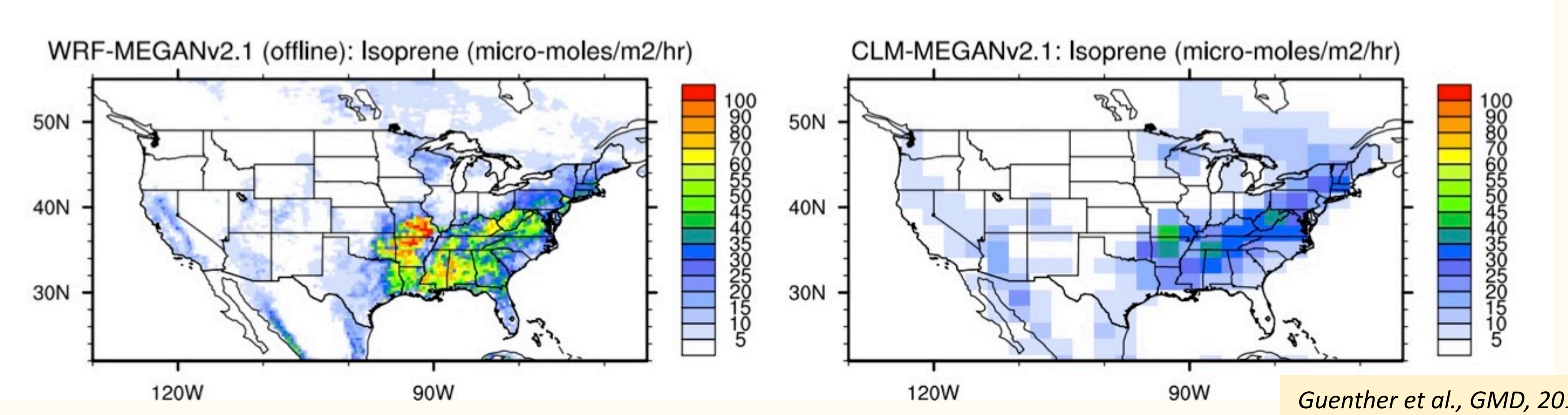
The MEGAN-v2.1 algorithm is online in many models

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, 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

Isoprene emissions in 2 different models differ greatly due to meteorology and vegetation maps



Guenther et al., GMD, 2012

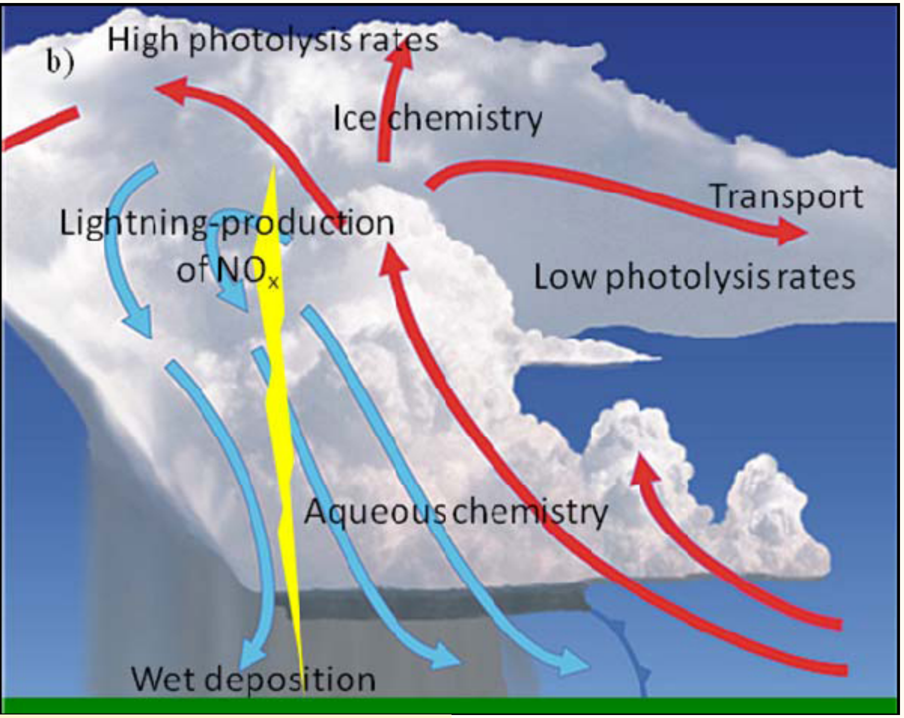
Lightning NO emissions

Lightning is produced in deep convective storms, creating a plasma leading to thermolysis of O_2 resulting in large NO emissions (1-10 Tg/yr globally)

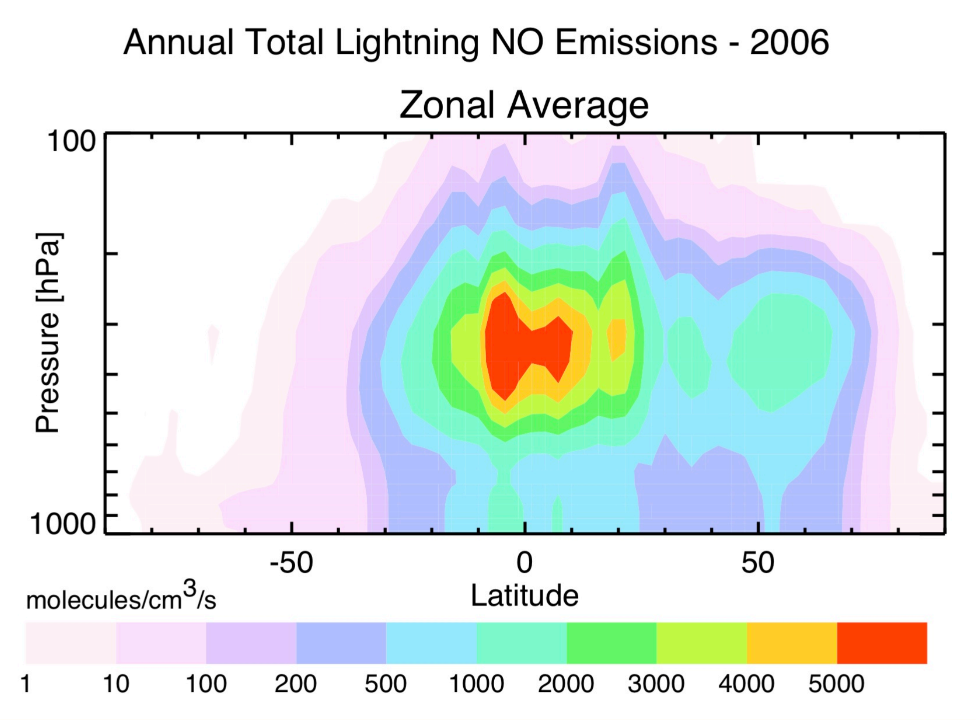
Often parameterized based on cloud top height

Cloud resolving models might use other approaches (e.g., updraft volume, ice mass flux, etc.

Can also use observations of lightning flashes

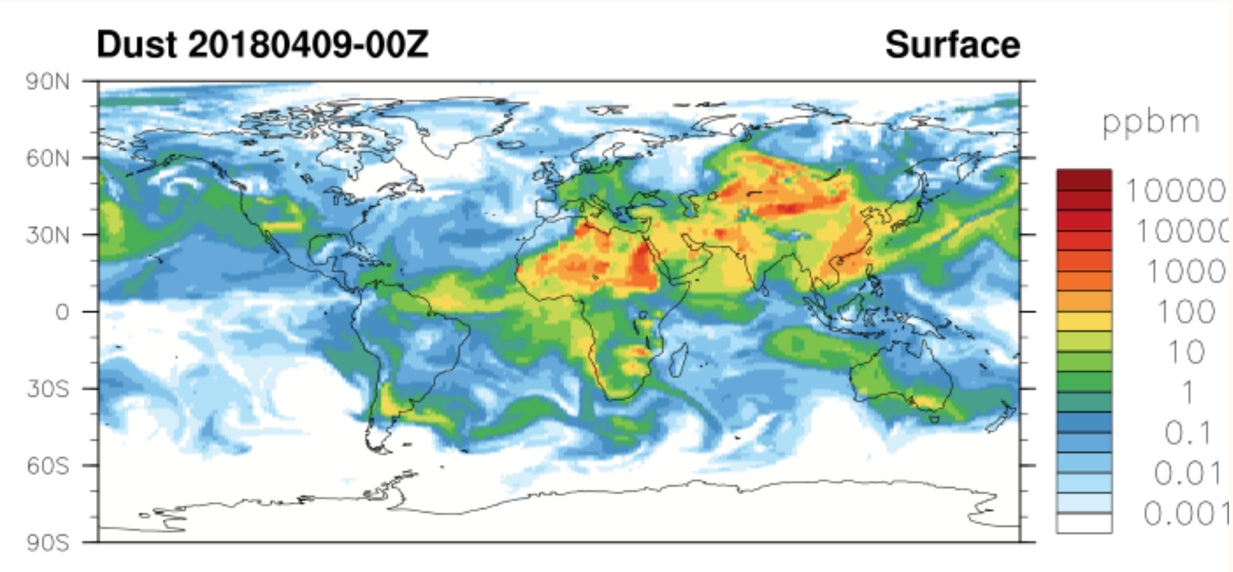


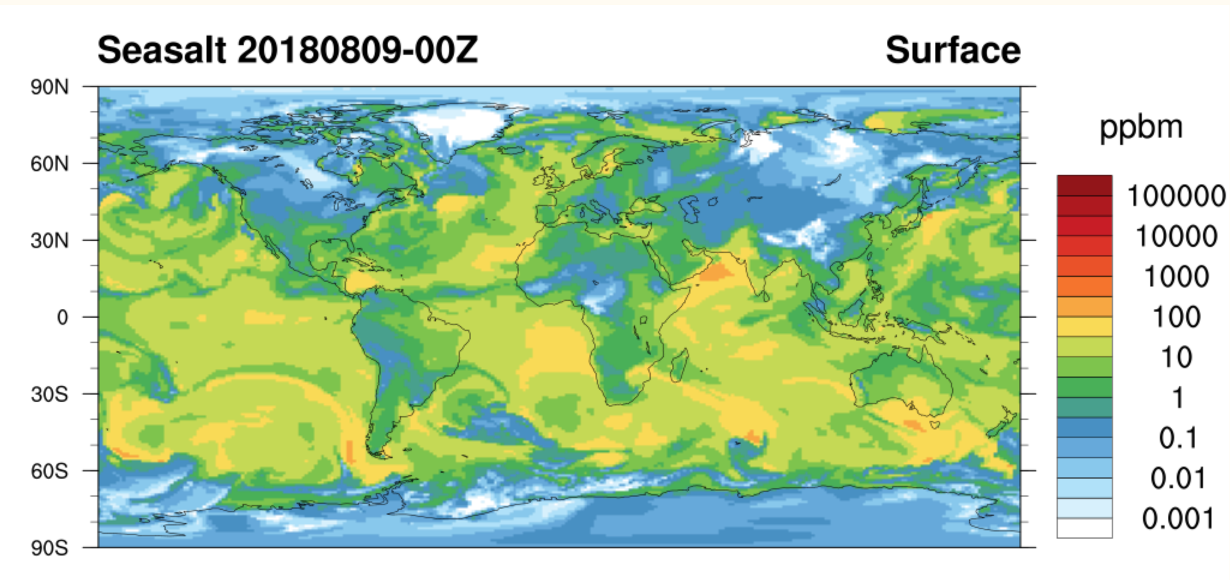
https://www2.acom.ucar.edu/dc3



Emmons et al., GMD, 2010

Dust and Sea salt





Parameterizations using surface wind speeds

Calculated online in model

Dust: based on soil erodibility maps, tuned to AOD observations

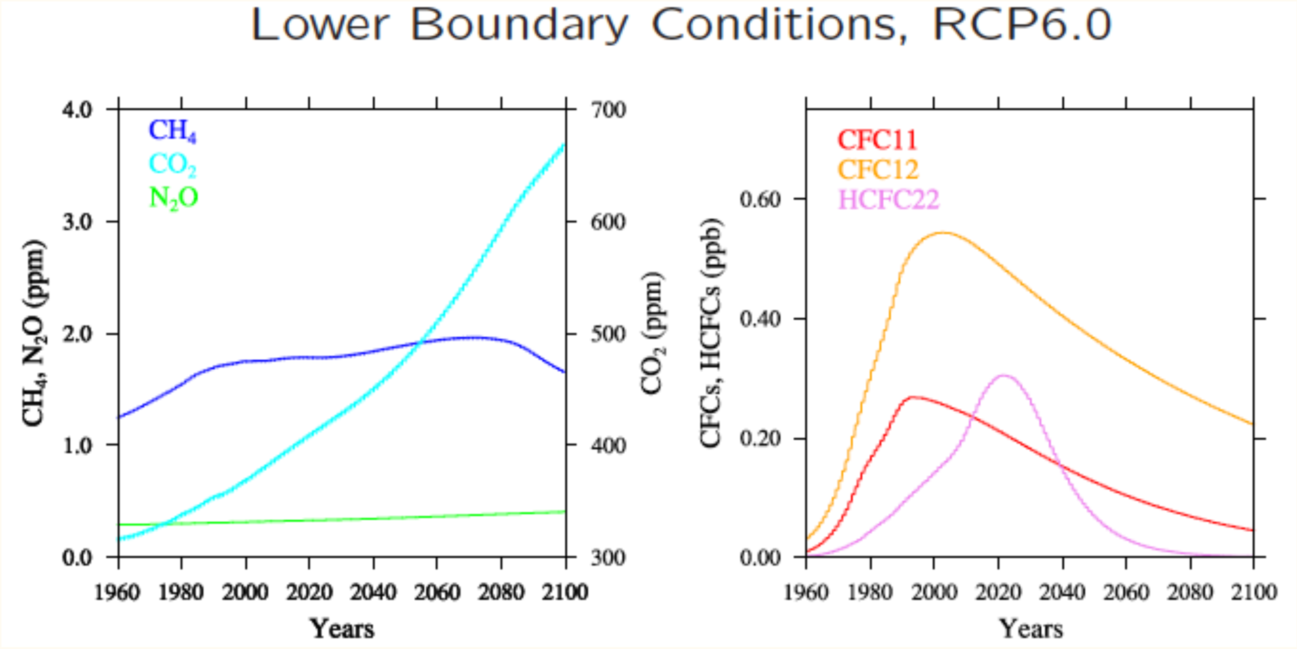
Images from CESM/WACCM: https://www.acom.ucar.edu/waccm/forecast/

Other Boundary Conditions

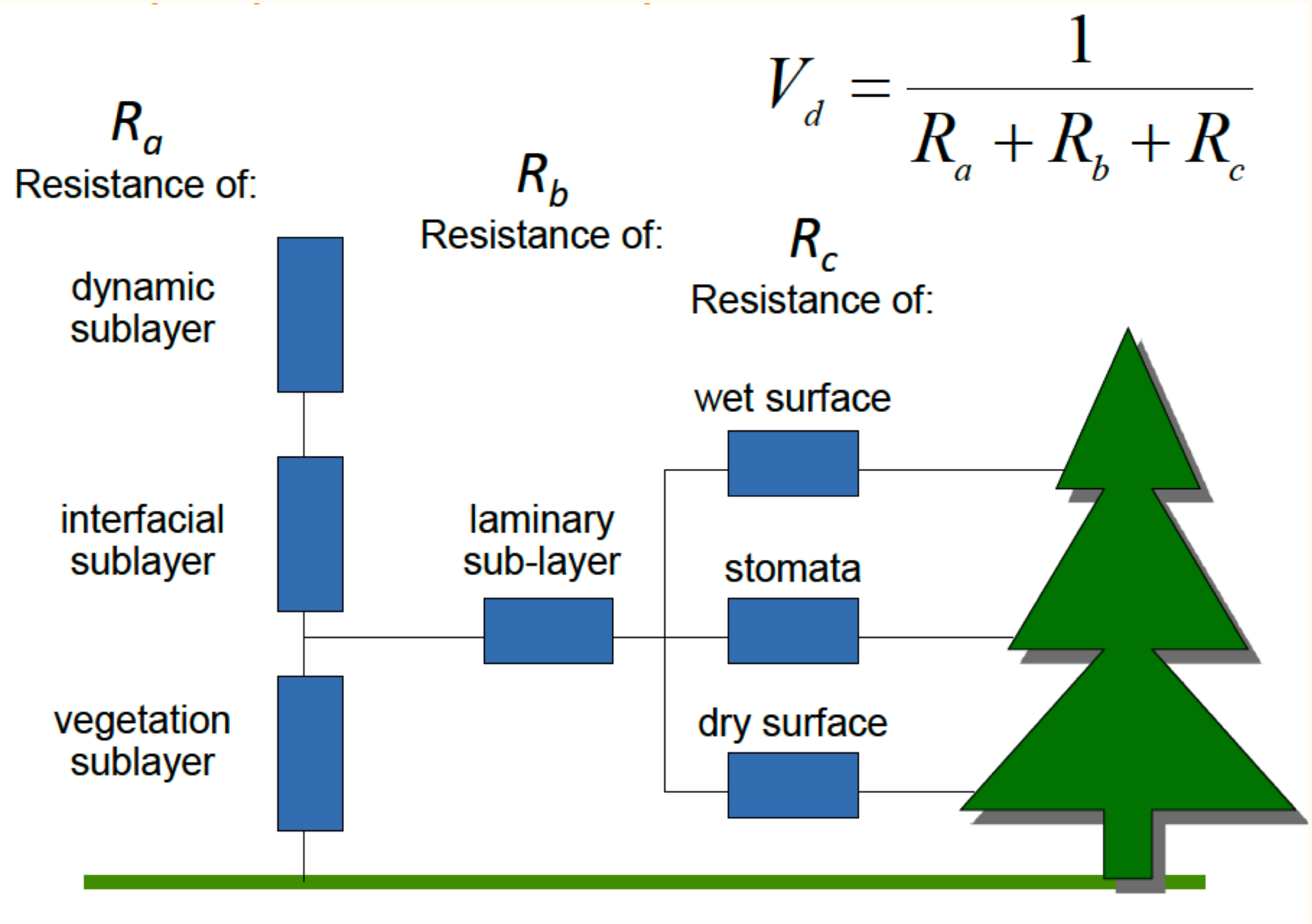
Long-lived species - for tropospheric chemistry often better to prescribe surface mixing ratios when sources and sinks are uncertain, such as for CH_4

Lateral boundary conditions for regional models

Stratosphere – ozone and other long-lived species (HNO3, NOx) are important source to troposphere



Dry Deposition Velocity



Varies with surface type (vegetation, ocean, etc.)
Key component of ozone budget
Important for sticky and soluble gases: HNO_3, CO, OVOCs, etc.

Wet Deposition

Wet deposition

- Scavenging in convective updrafts
- Rainout: in-cloud scavenging
- Washout: below-cloud scavenging

Highly soluble gases (HNO_3) and aerosols - kinetic process limited by mass transfer

Moderately soluble gases: dependent on effective Henry's Law constant

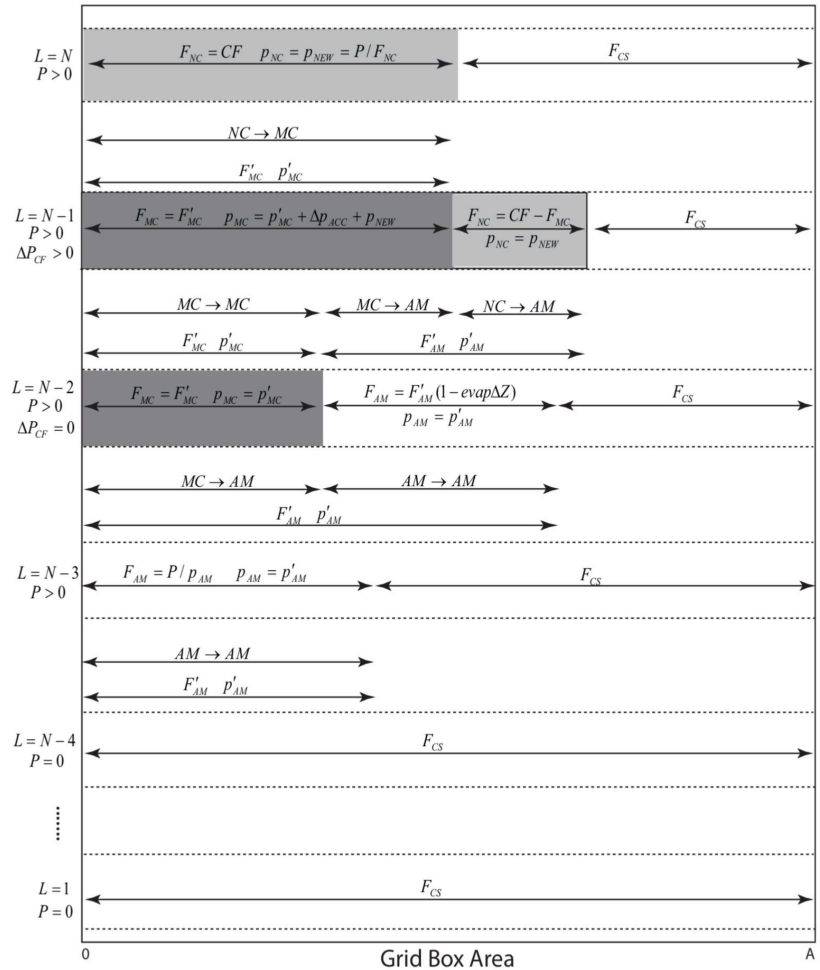
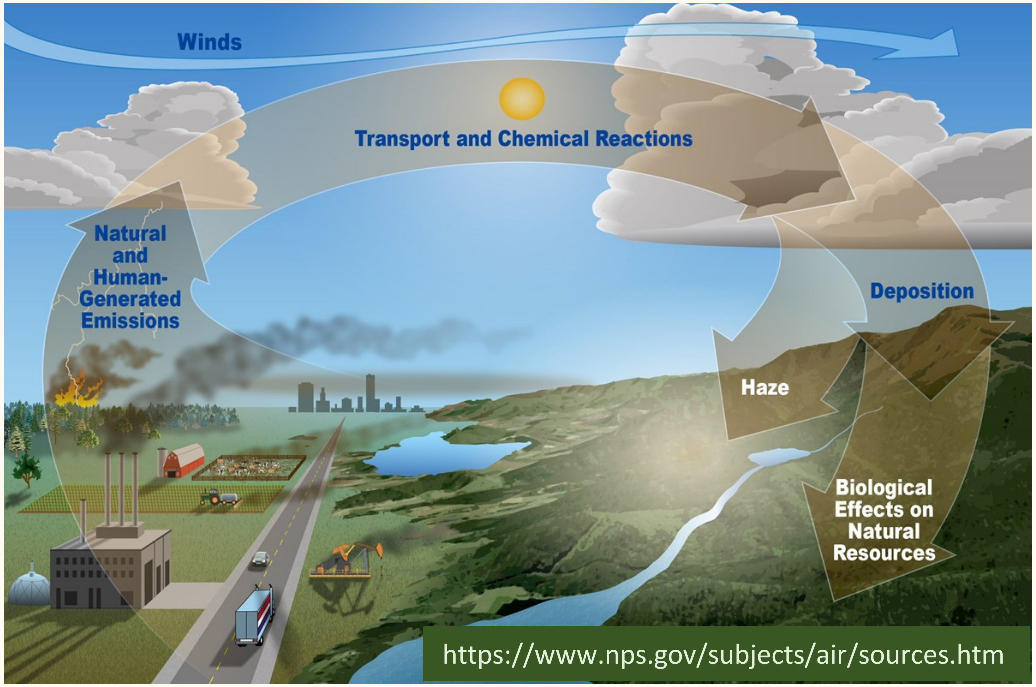
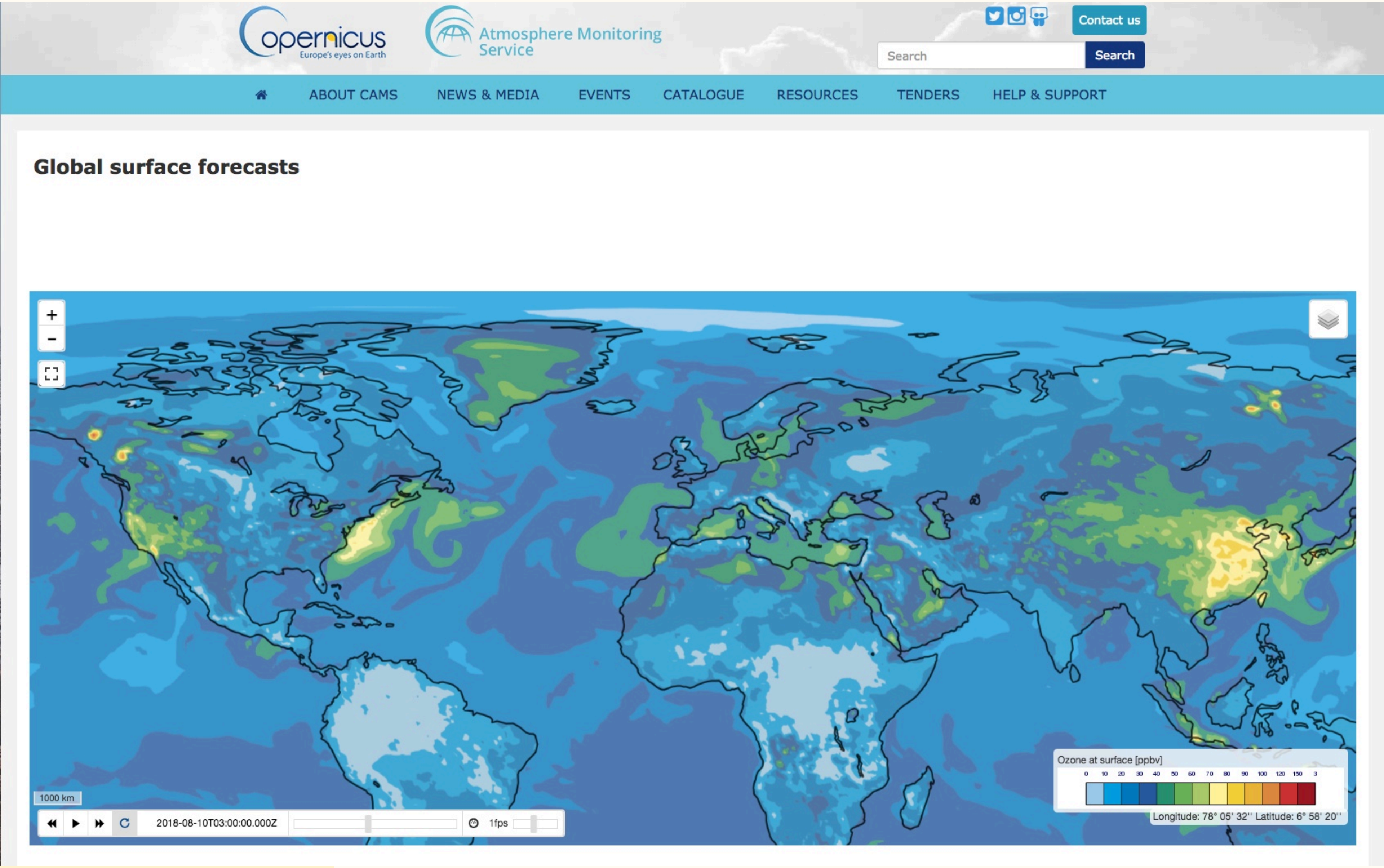


Fig. A1. Schematic of an idealized grid box with N levels, indicated by the horizontal dotted lines. The space between the levels is for illustrative purposes only. Levels N to $N-2$ contain clouds (grey shaded areas) and Levels N to $N-3$ have $P(L) > 0$. All gridbox fractions and precipitation rates are defined in the text and are shown for each level. The terms XX → YY indicate the gridbox fraction corresponding to $\hat{f}_{\mathrm{XX}\rightarrow\mathrm{YY}}$ as defined in the text (Eqs. A3 to A8). Level N is the first precipitating level, and the precipitating fraction and rate are given by Eqs. (A1) and (A21). Level $N-1$ provides an example with $CF(L) > CF(L+1)$, $\Delta P_{CF} > 0$ (Eq. A18), and $p_{\mathrm{NEW}} > 0$ (p_{MC} and p_{NC} given by Eqs. A27 and A28). Level $N-2$ has $CF(L+1) > CF(L)$ and $\Delta P_{CF} = 0$, with the standard evaporation in the ambient region sufficient to account for the decrease in P from level $N-1$ to $N-2$ (Eqs. A16 and A17). Level $N-3$ has $CF(L) = 0$ and the decrease in P from level $N-2$ to $N-3$ exceeds the standard evaporation rate (Eq. A17). There is full evaporation of the precipitation in level $N-4$ ($P(L) = 0$).

Applications

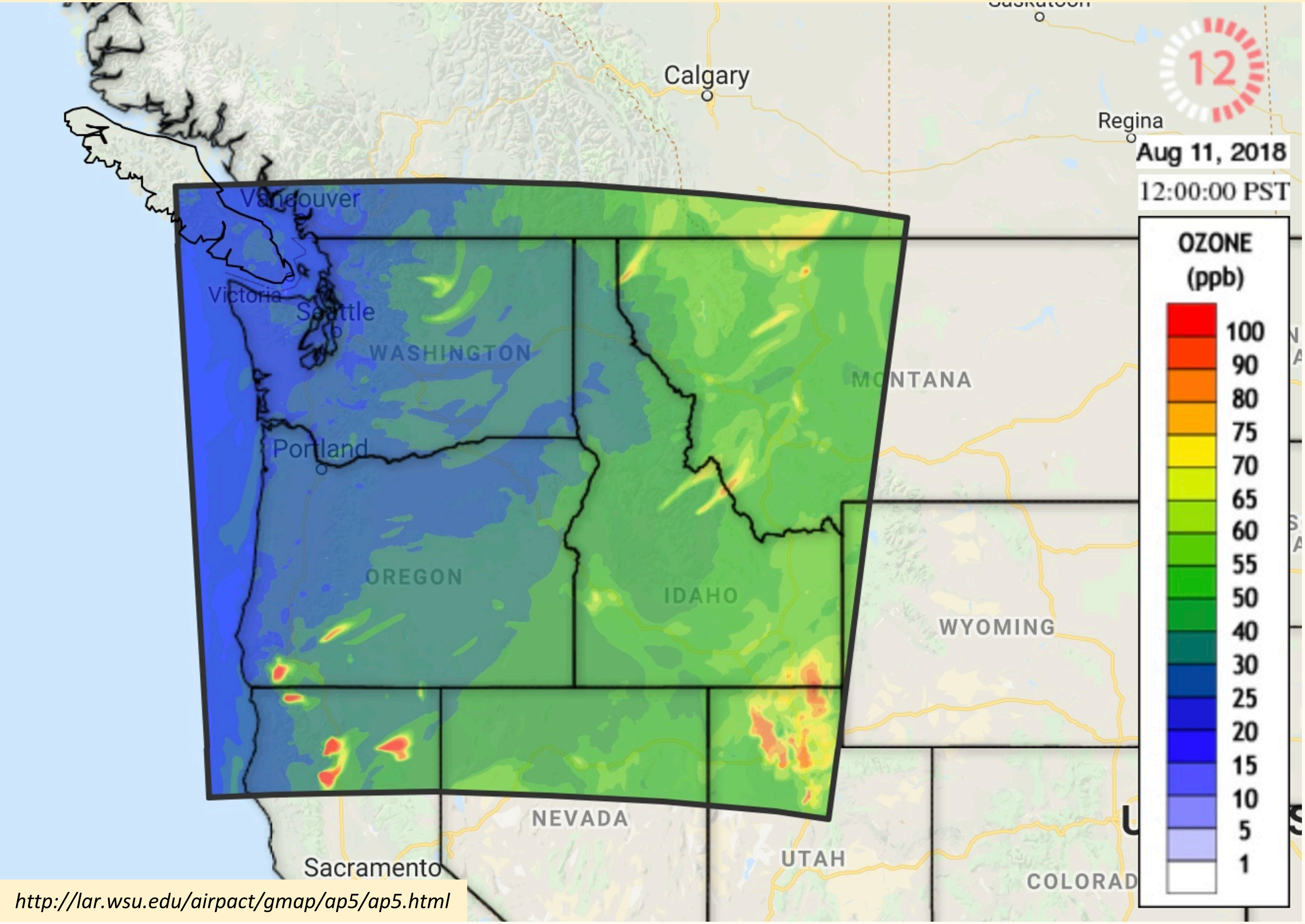
- Chemical & air quality forecasts
- Source attribution through “tagging”
- Analysis of observations and understanding processes
- Chemistry-climate coupling





https://atmosphere.copernicus.eu/

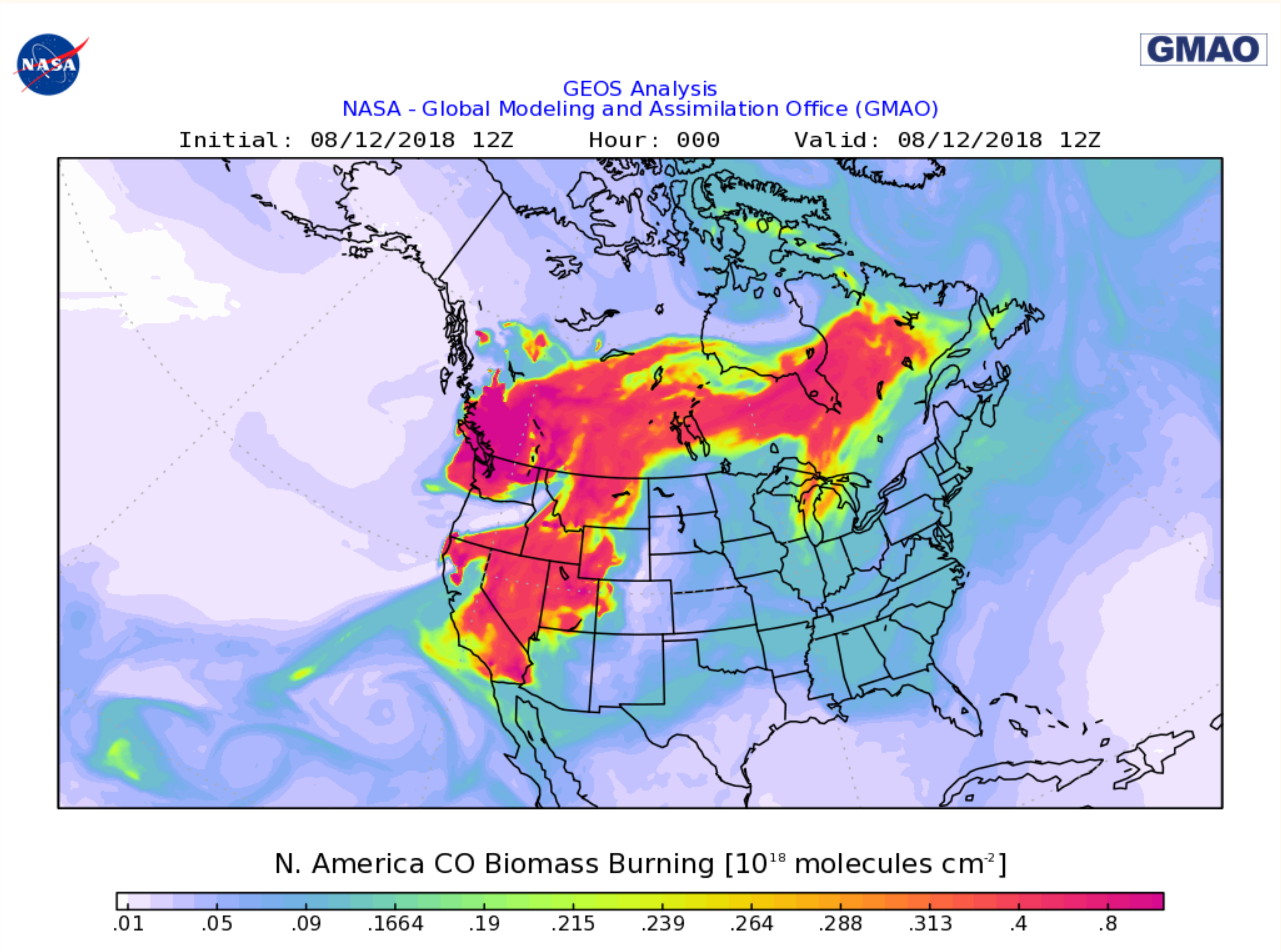
Regional AQ forecasts (e.g., AIRPACT, WSU)



http://lar.wsu.edu/airpact/gmap/ap5/ap5.html

Source Attribution

”Tagging” of source type or region allows identification of source contributions to observations



Frequently used in flight planning for aircraft experiments

https://fluid.nccs.nasa.gov/wxmaps/chem2d/

Adding tagging to chemical mechanisms

Artificial tracers:
CO_fire_10day -> CO2 : k=1/10days, CO emissions from fires

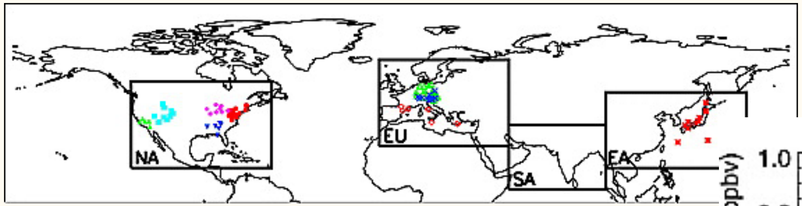
SO2_volcano -> {} : k=1/x, SO_2 emissions from active volcano

”Tagged” CO, runs parallel to complete chemistry without affecting it:
CO_fire + OH -> OH : k=k(CO+OH)

Other compounds with simple chemistry can be tagged and added to standard chemistry (BC)

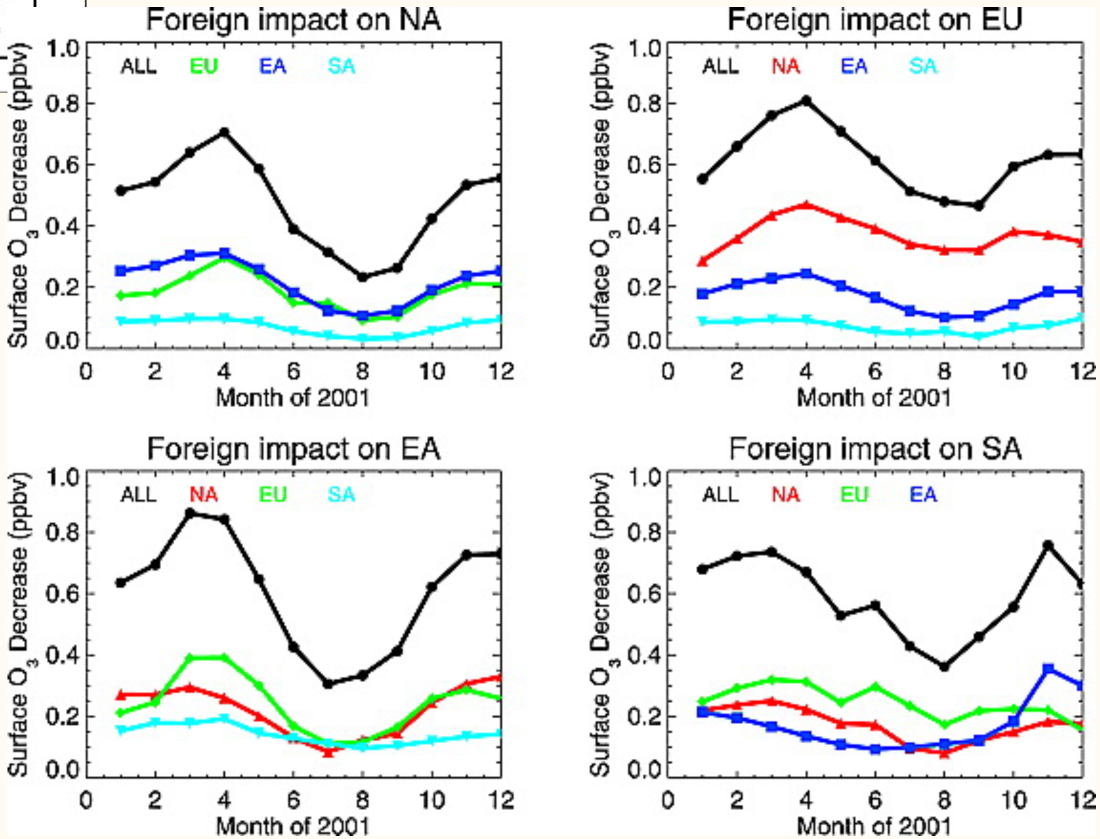
Emissions perturbations

- For studying impacts of actually changing the emissions
- To estimate source contributions from various source types or regions



HTAP model intercomparison

Decreased emissions in each region by 20% and found decrease in ozone



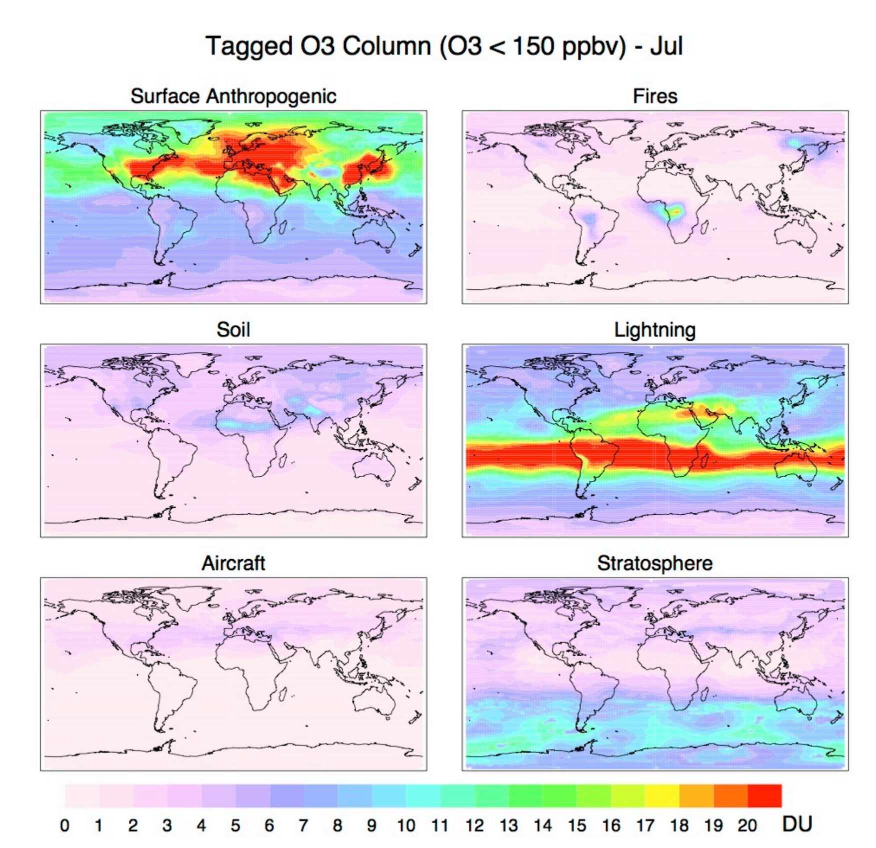
Fiore et al., JGR, 2009

Tracking ozone from NO emissions

$NO_2 + h\nu \rightarrow NO + O$	$XNO_2 + h\nu \rightarrow XNO + OA$
$O + O_2 \rightarrow O_3$	$OA + O_2 \rightarrow O3A$
$NO + O_3 \rightarrow NO_2 + O_2$	$XNO + O_3 \rightarrow XNO_2$
	$NO + O3A \rightarrow NO$
$NO_2 + OH \rightarrow HNO_3$	$XNO_2 + OH \rightarrow XHNO_3 + OH$
$HNO_3 + OH \rightarrow NO_3 + H_2O$	$XHNO_3 + OH \rightarrow XNO_3 + OH$

Any region or source type of NO emissions can be tagged, to quantify contribution of ozone from that source

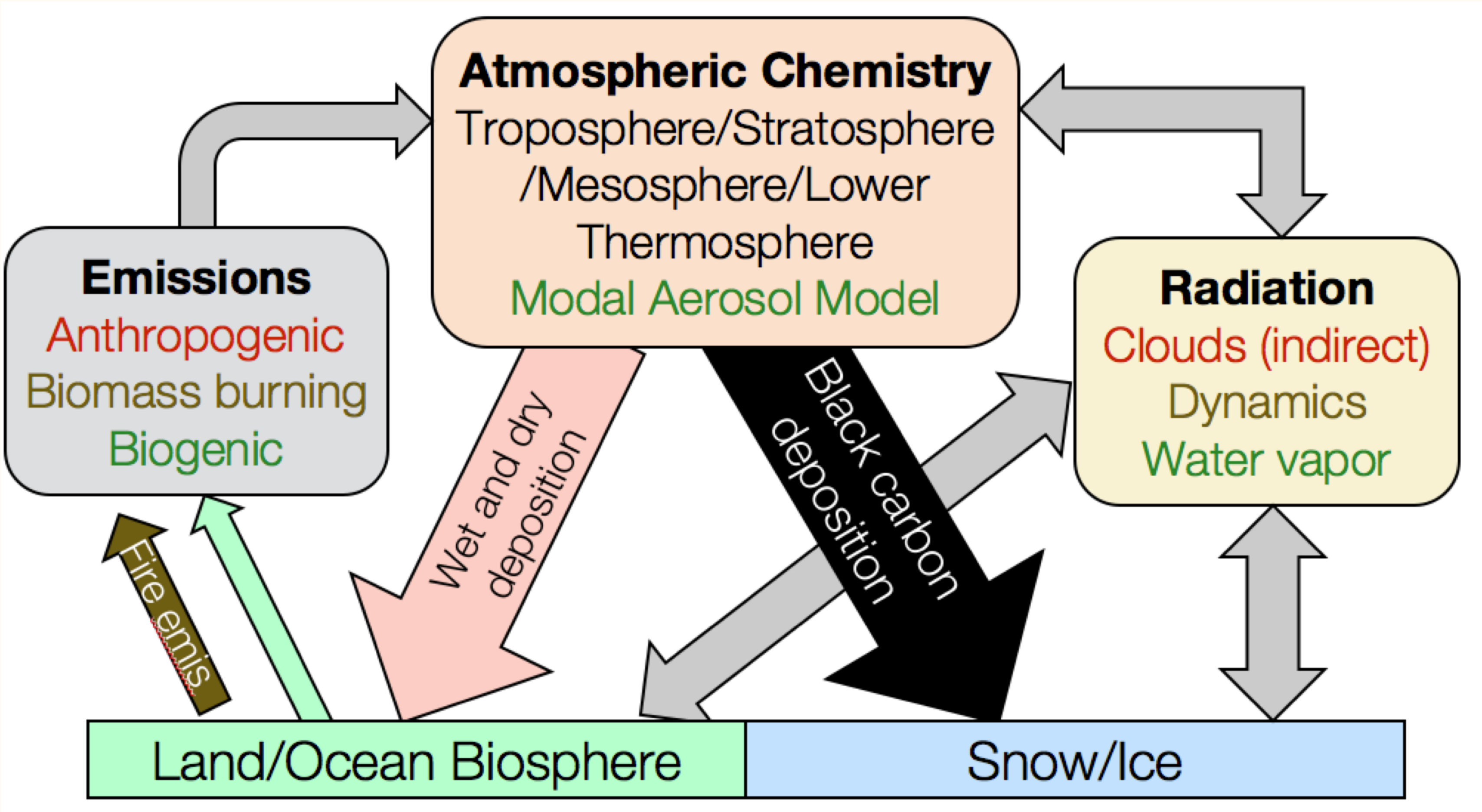
Similar tagging of VOCs -> ozone also possible (Butler, GMD, 2018)



Emmons et al., GMD, 2012

Aircraft experiments: forecasting and analysis

Chemistry-Climate Interactions



Conclusions

- Models are tools to explore atmospheric processes
- Emissions are very uncertain and should be appropriately selected and evaluated for each study
- Uncertainties in other processes (deposition, chemistry, clouds, meteorology) must be kept in mind