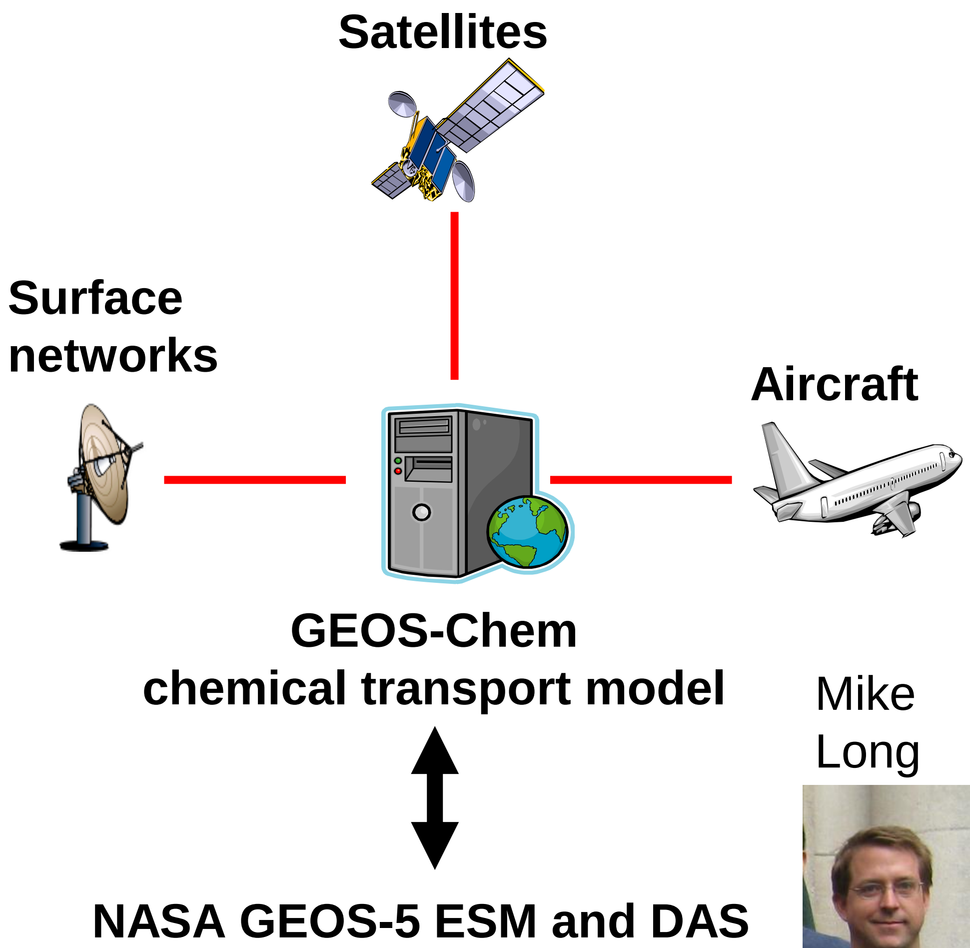
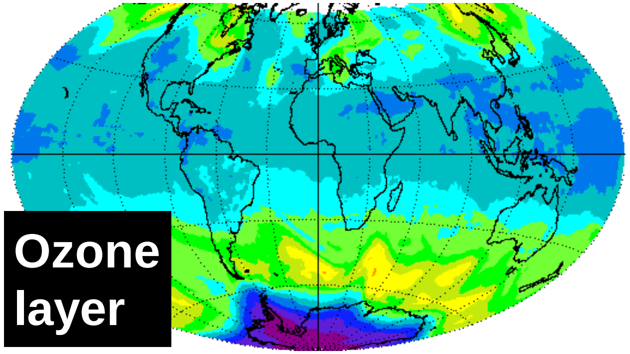
The role of atmospheric composition in Earth system modeling for numerical weather production

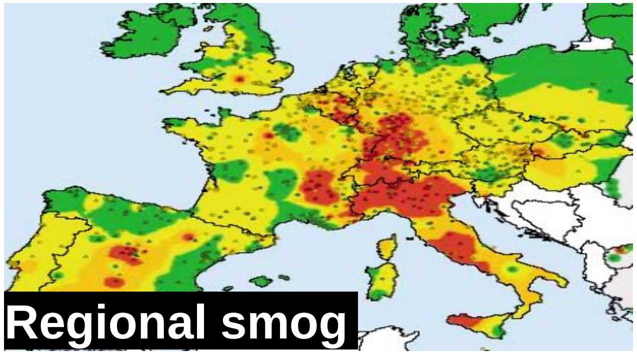
Daniel J. Jacob

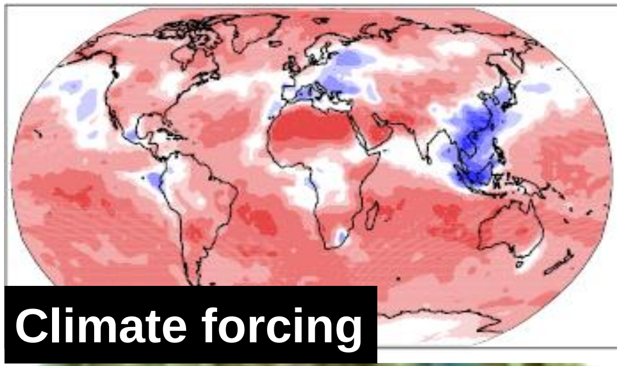
HARVARD UNIVERSITY
ATMOSPHERIC CHEMISTRY MODELING GROUP

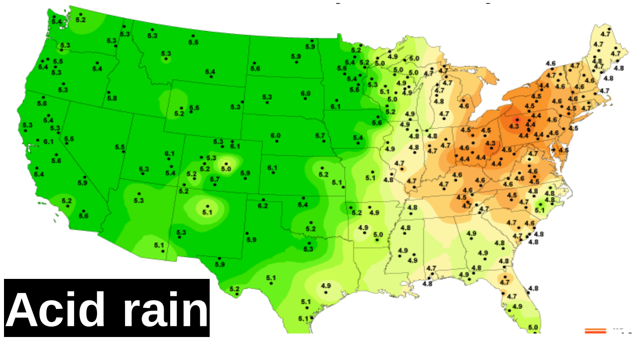


Reasons to care about atmospheric chemistry









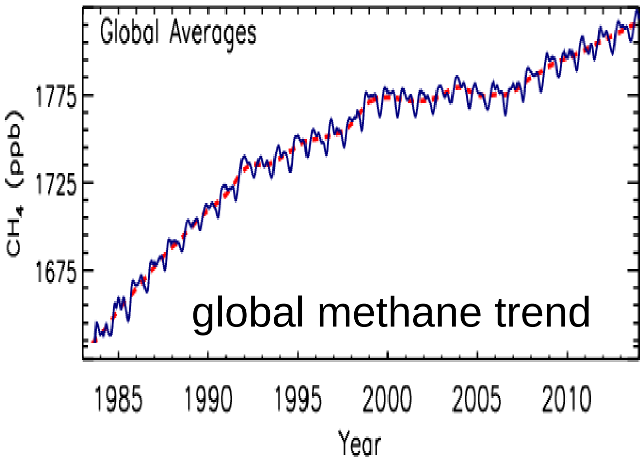
LOCAL
< 100 km

REGIONAL
100-1000 km

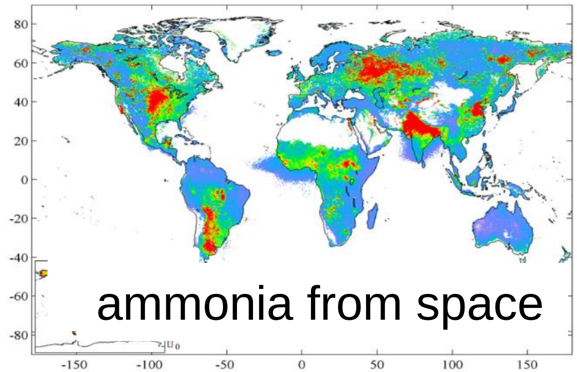
GLOBAL
> 1000 km

Some big questions for tropospheric chemistry in next decade

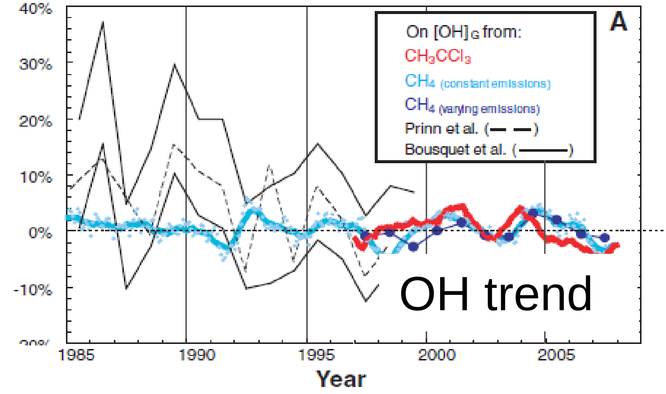
Changing methane



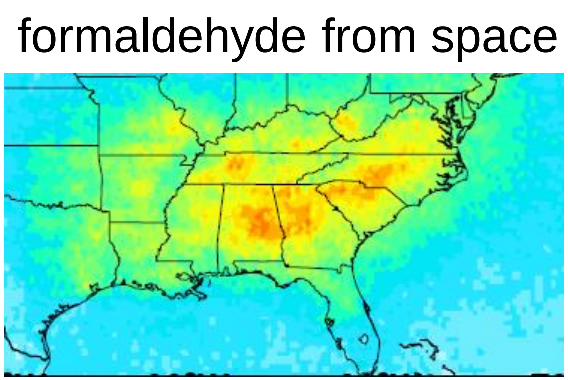
Changing N cycle



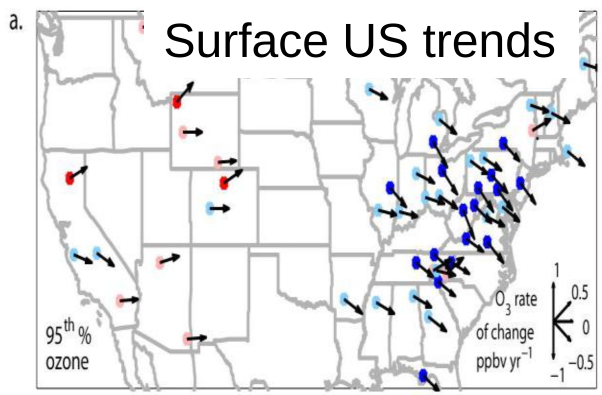
Trends in oxidants



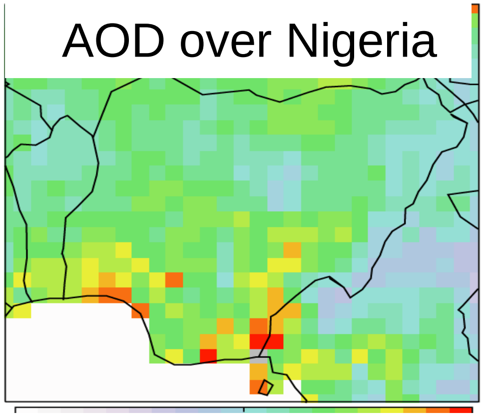
Biogenic organics



Ozone trends

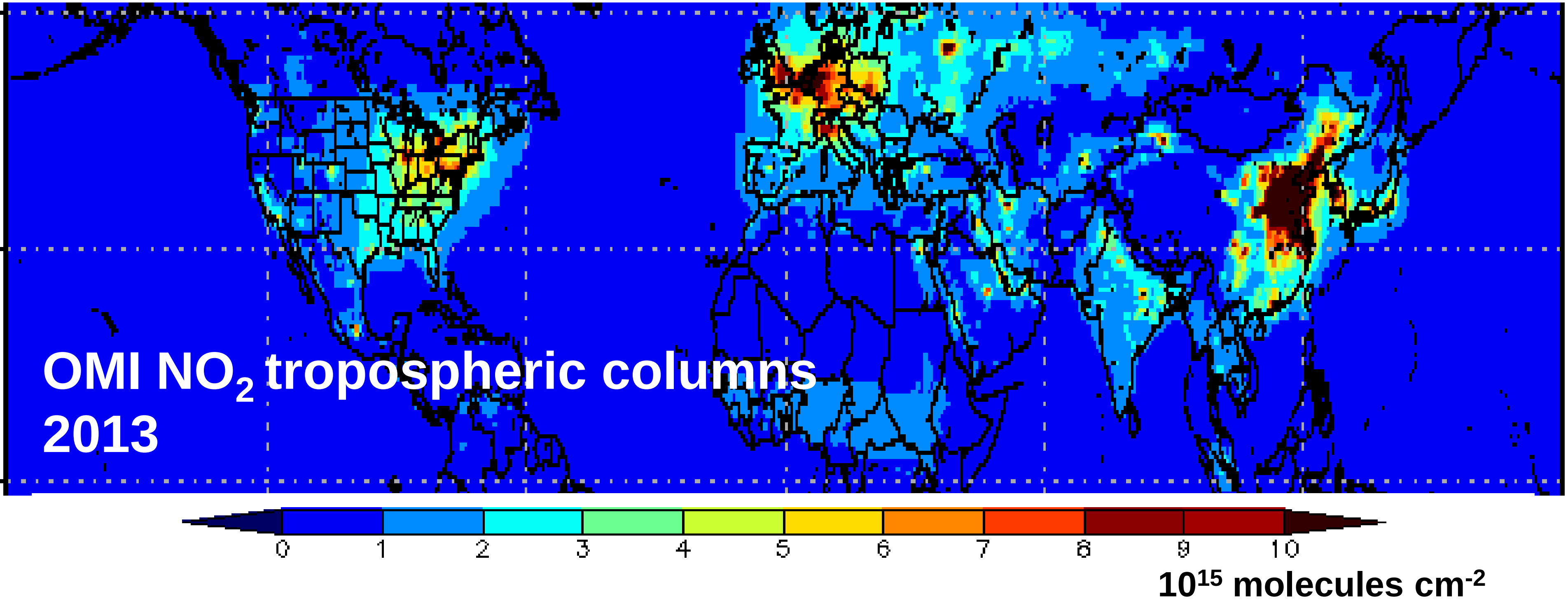


Air quality in developing world



Emerging era of satellite observations

Tropospheric chemistry has transitioned from data-poor to data-rich over past 15 years



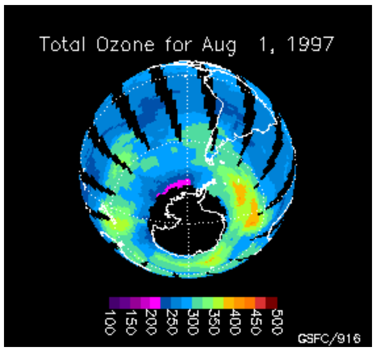
This has provided impetus for development of chemical data assimilation tools

- Inverse analyses of emissions
- Chemical reanalyses
- Initialization of chemical forecasts
- Improvement of meteorological forecasts

Improving meteorological forecasts through chemical information

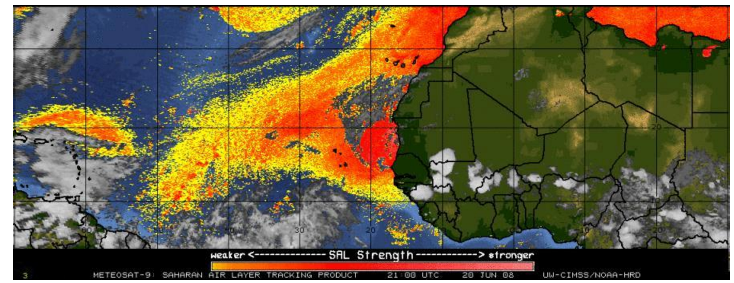
Ozone for stratospheric dynamics

Ozone columns, profiles



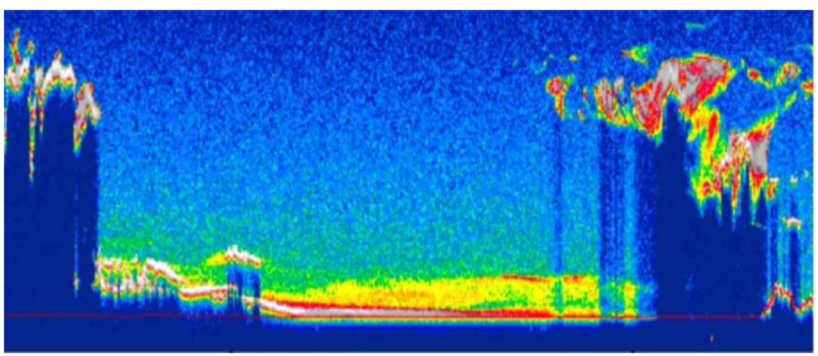
Aerosols for radiation/precipitation

GOES aerosol optical depth



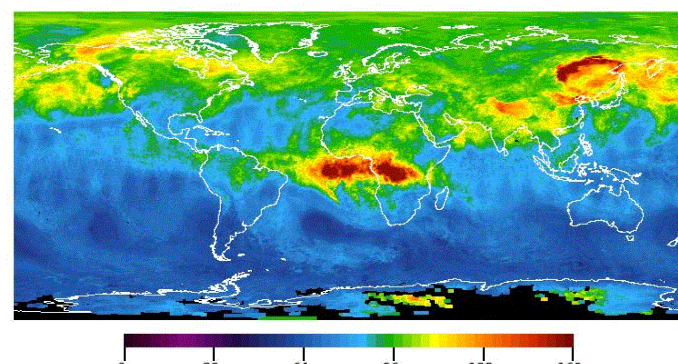
PBL heights

CALIOP lidar aerosol profiles



Chemical tracers of winds

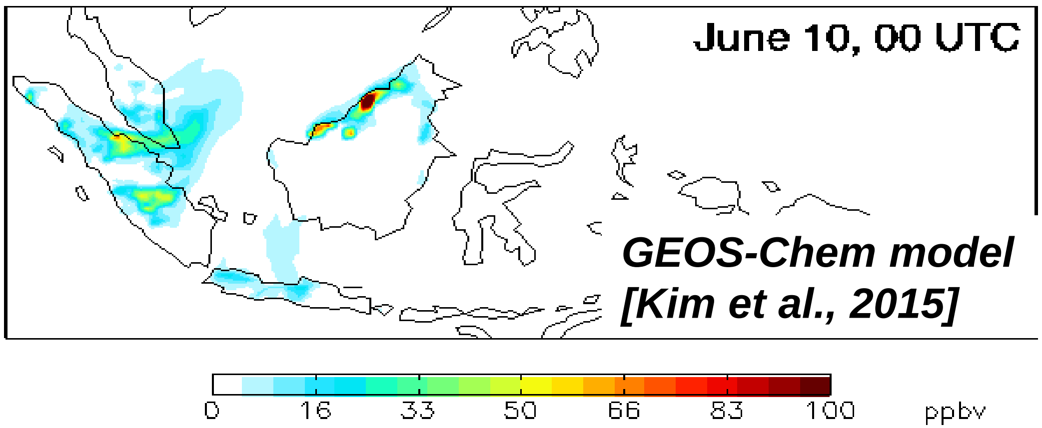
Free tropospheric carbon monoxide (CO)

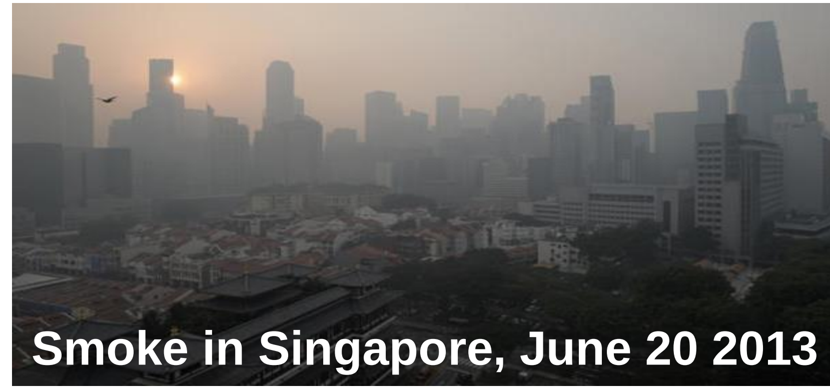


Public demand for chemical forecasts

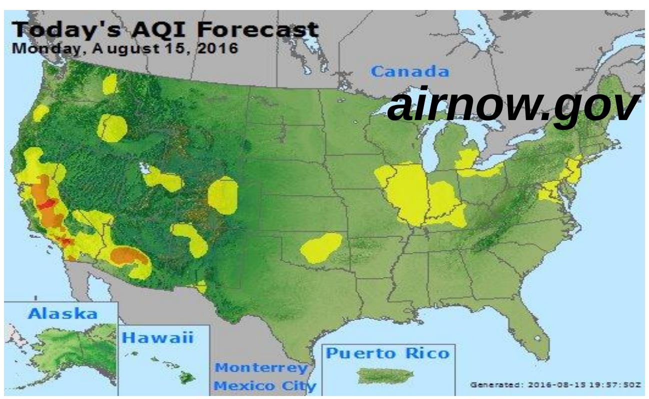
Transport of pollution from major point releases (fires, volcanoes, accidents)

Smoke from agricultural fires in Sumatra

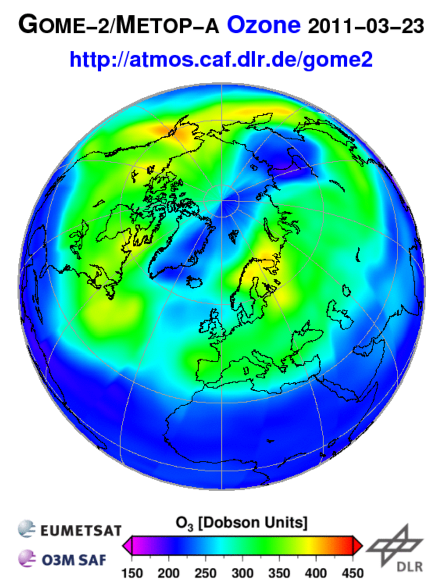




Air quality management

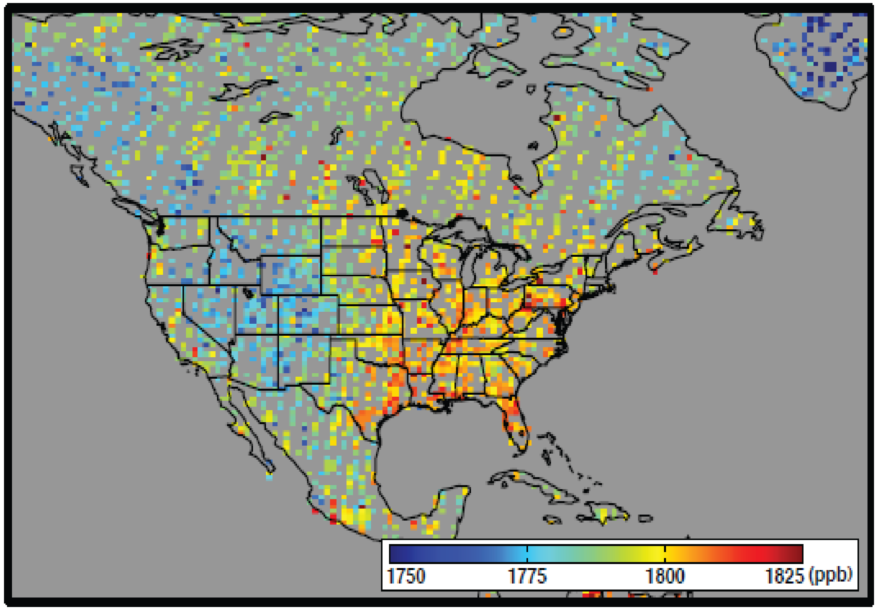


Ozone hole drift

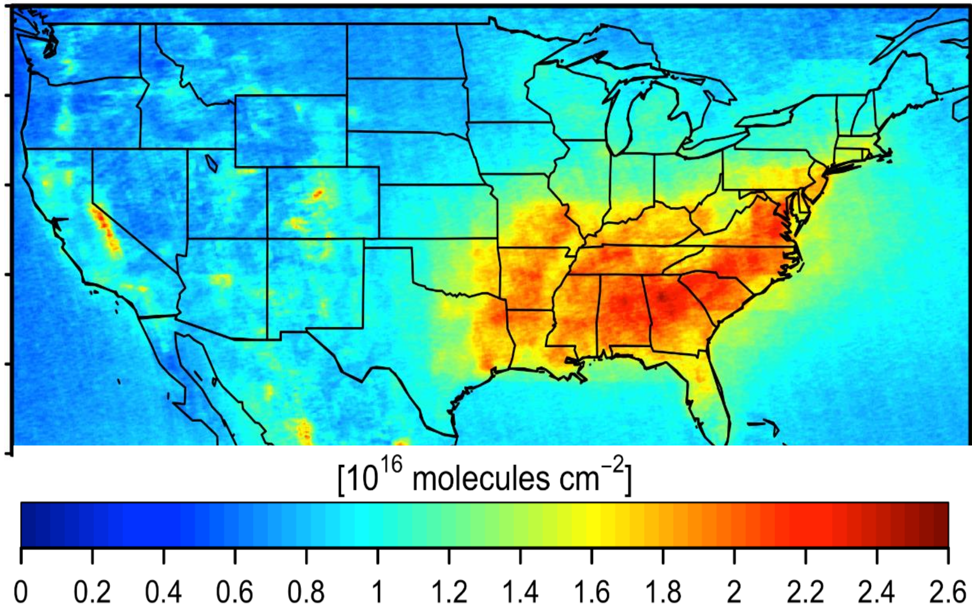


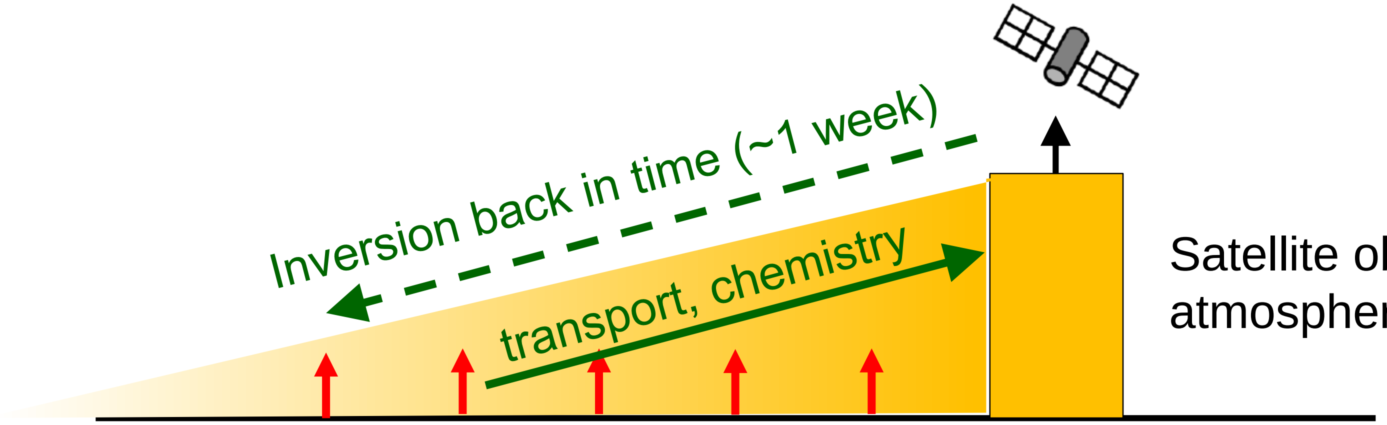
Monitoring emissions in near-real-time from satellite data

GOSAT methane: methane emissions



OMI formaldehyde: hydrocarbon emissions





Computational cost of chemical models

Solve n coupled PDEs for species mixing ratio $\mathbf{C} = (C_1, ..., C_i, ... C_n)^T$

$$\partial C_i / \partial t = \underbrace{-\mathbf{u}\nabla C_i}_{\text{advection}} + \underbrace{\nabla K \nabla C_i}_{\text{subgrid}} + \underbrace{P_i(\mathbf{C})}_{\text{production}} - \underbrace{L_i(\mathbf{C})}_{\text{loss}} + \underbrace{E_i}_{\text{emission}} - \underbrace{D_i}_{\text{deposition}}$$

operator splitting

Transport modules: PDEs with no coupling across species

$$\partial C_i / \partial t = -\mathbf{u}\nabla C_i + \nabla K \nabla C_i$$

Chemical module: stiff coupled system of ODEs.

$$dC_i / dt = P_i(\mathbf{C}) - L_i(\mathbf{C}) + E_i - D_i$$

For a typical mechanism with ~100 coupled species, chemical module is expensive!

But:

- There are fast implicit solvers available for such stiff systems
- The chemical module has 100% scaling in massively parallel environments
- Chemistry may use coarser time steps and grid resolution than dynamics
- As grid resolution increases, cost of chemistry vs. transport decreases
- Mechanism may be reduced in clean regions

On-line and off-line approaches to chemical modeling

On-line: coupled to dynamics

GCM conservation equations:
air mass: $\partial \rho_a / \partial t = \ldots$
momentum: $\partial \mathbf{u} / \partial t = \ldots$
heat: $\partial \theta / \partial t = \ldots$
water: $\partial q / \partial t = \ldots$
chemicals: $\partial C_i / \partial t = \ldots$

Off-line: decoupled from dynamics

GCM conservation equations:
air mass: $\partial \rho_a / \partial t = \ldots$
momentum: $\partial \mathbf{u} / \partial t = \ldots$
heat: $\partial \theta / \partial t = \ldots$
water: $\partial q / \partial t = \ldots$

↓

meteorological archive
(averaging time ~ hours)

↓

Chemical transport model:
$\partial C_i / \partial t = \ldots$

PROs of off-line vs on-line approach:
- computational cost
- simplicity
- stability (no chaos)
- compute sensitivities back in time

CONs:
- no chemical-dynamics coupling
- need for meteorological archive
- transport errors

Chemical data assimilation best done on-line ↔ **Chemical sensitivity studies may best be done off-line**

GEOS-Chem Chemical Transport Model:

off-line model using NASA GEOS operational meteorological archive

Input data

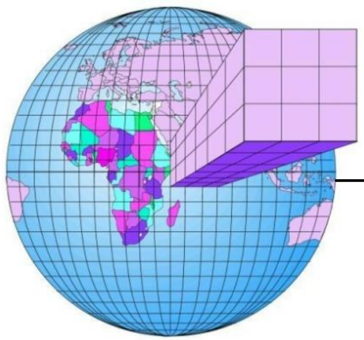
NASA GEOS-5 meteorological fields:

0.25°x0.3125° horizontal resolution, 72 vertical levels up to 0.1 hPa

GEOS-Chem solves 3-D chemical continuity equations
on global or nested Eulerian grid

Modules

- emissions
- transport
- chemistry
- aerosols
- deposition
- sub-surface



Model adjoint

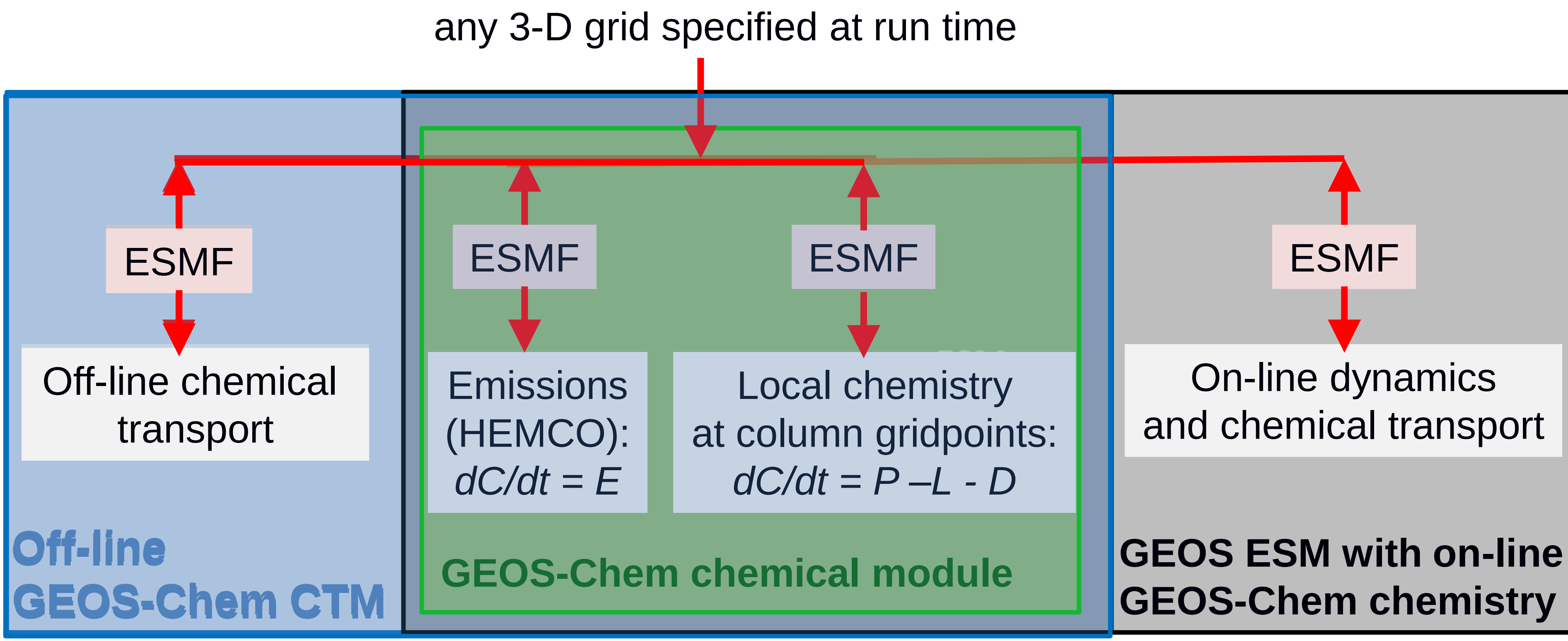
Applications

- chemical, aerosol processes
- inversions of surface fluxes
- radiative forcing
- air quality
- biogeochemistry
- …

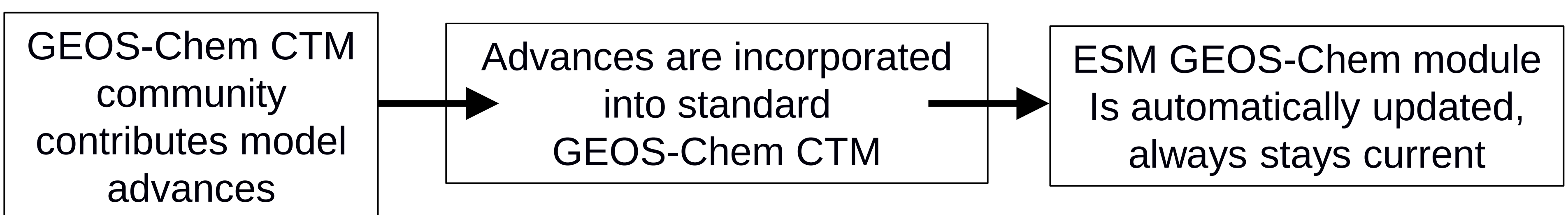
Developed and used by over 100 research groups worldwide

GEOS-Chem chemical module can be used off-line or on-line

grid-independent modules connected by Earth System Modeling Framework (ESMF)



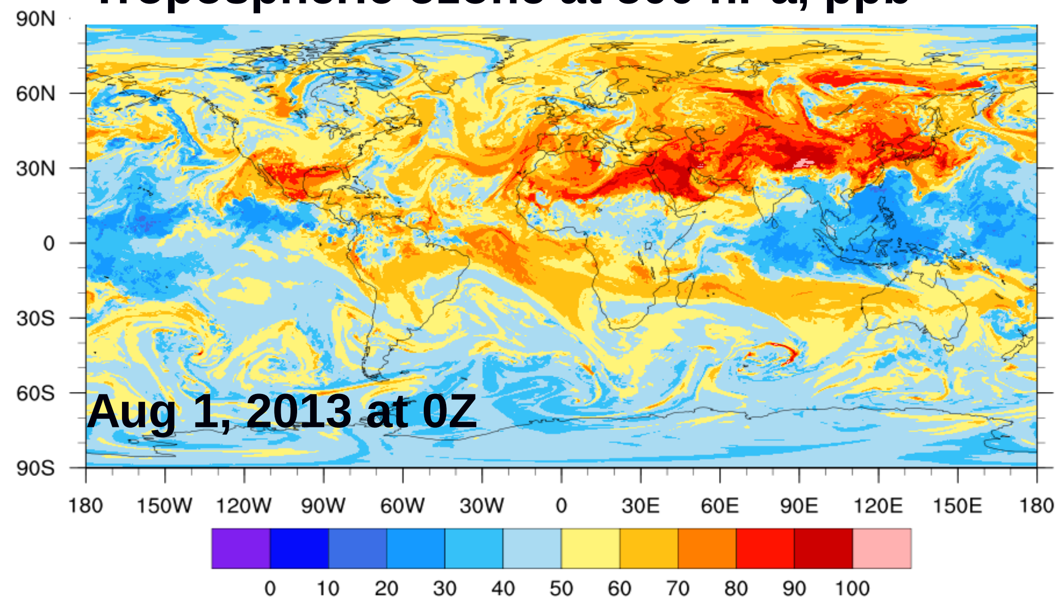
GEOS-Chem chemical module in CTM and ESM is exactly the same code



Long et al. [2015]

GEOS-Chem chemistry in c720 (12 km) GEOS-5 ESM

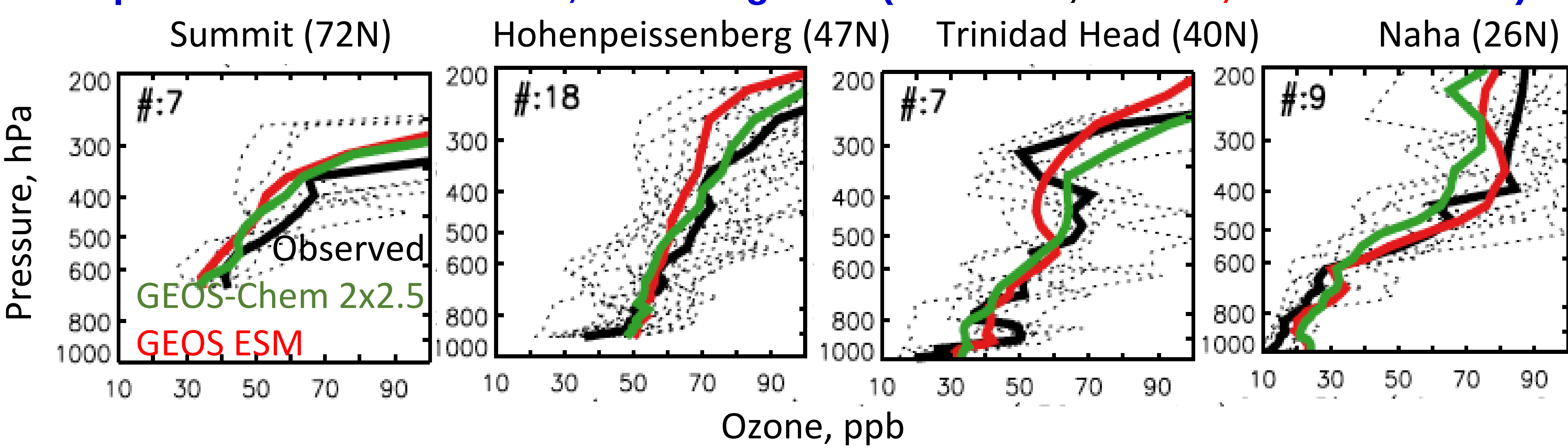
Tropospheric ozone at 500 hPa, ppb



Full-year simulation:

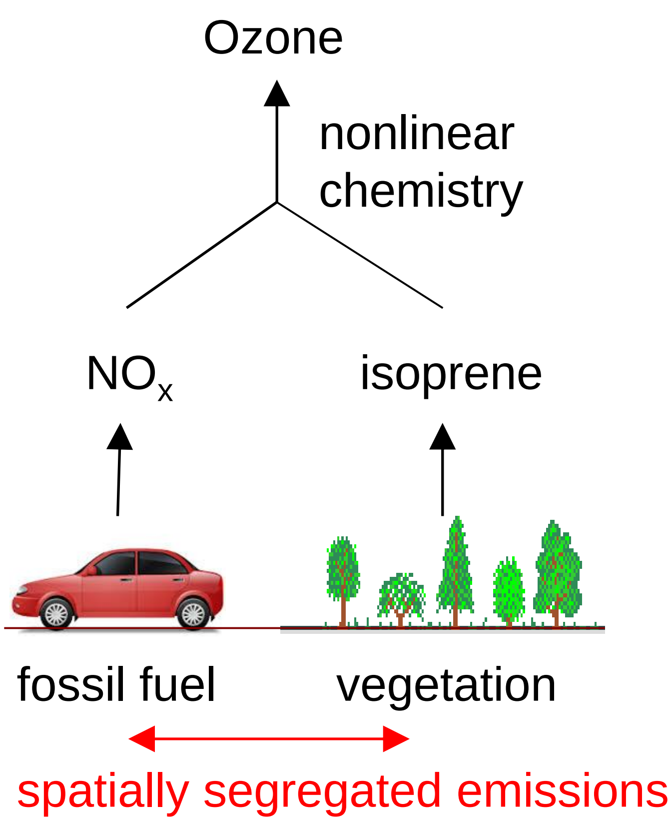
Mike Long, Lu Hu (Harvard), Christoph Keller (NASA)

Comparison to ozonesondes, June-Aug 2013 (observed, on-line, off-line 2°x2.5°)

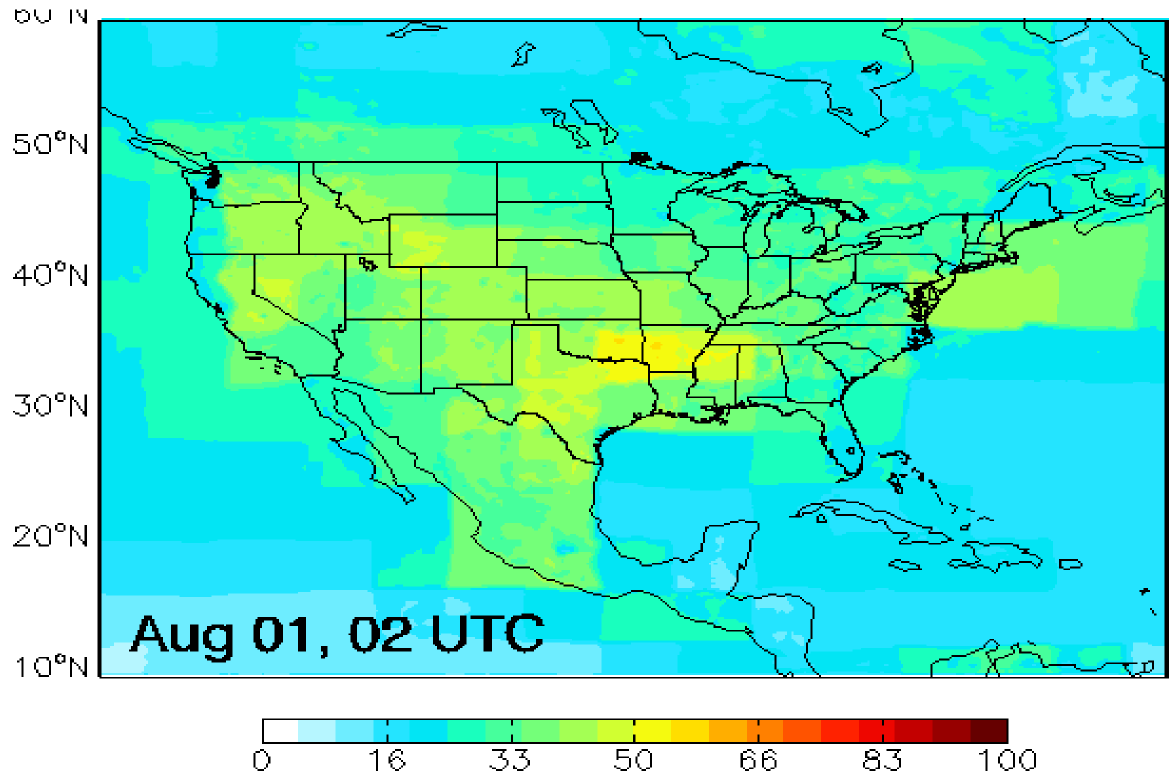


Nonlinear chemistry and grid resolution

GEOS-Chem with 0.25°x0.3125° resolution over North America
during NASA SEAC4RS aircraft campaign over Southeast US (Aug-Sep 2013)
5-min transport time step, 10-min chemical time step (*h*)



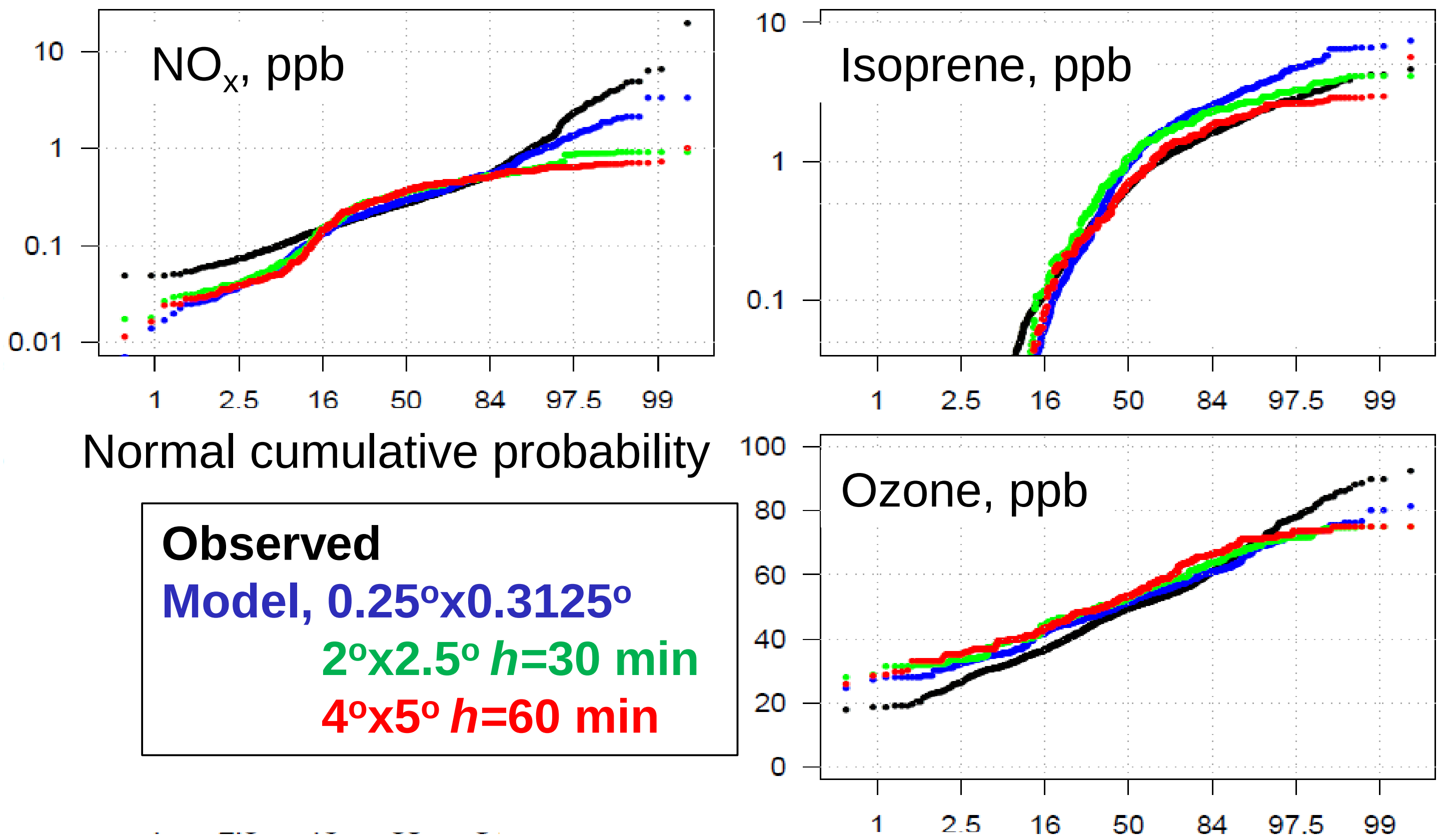
Oxone in surface air - circles are aircraft data



Yu et al. [ACP 2016]

Effect of grid resolution on nonlinear chemistry is small

Cunulative PDFs of observations and model (different resolutions) over Southeast US

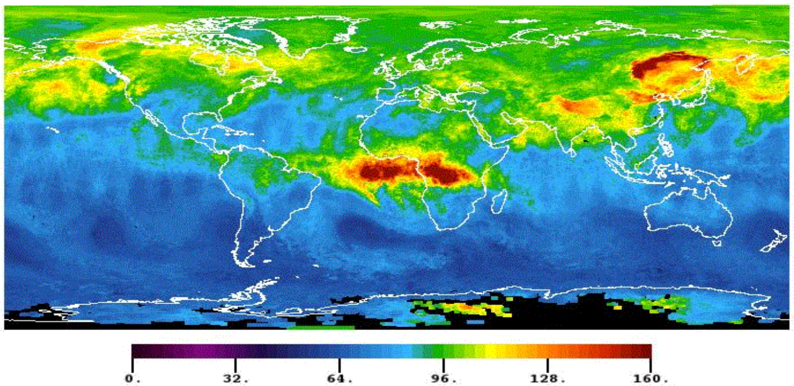


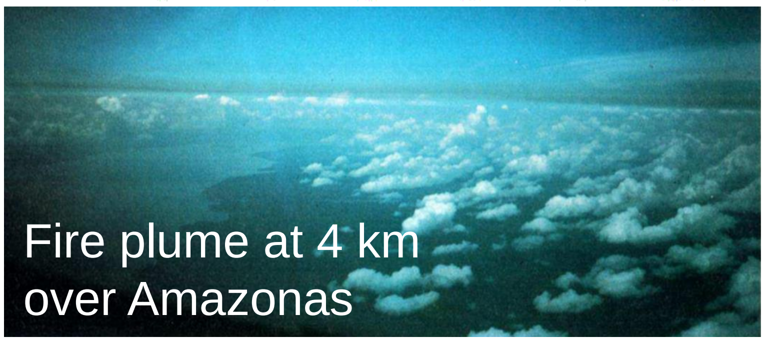
Chemical averaging errors tend to elicit negative feedback (LeChatelier principle): a high-resolution dynamical model could use coarser resolution for chemistry

Yu et al. [ACP 2016]

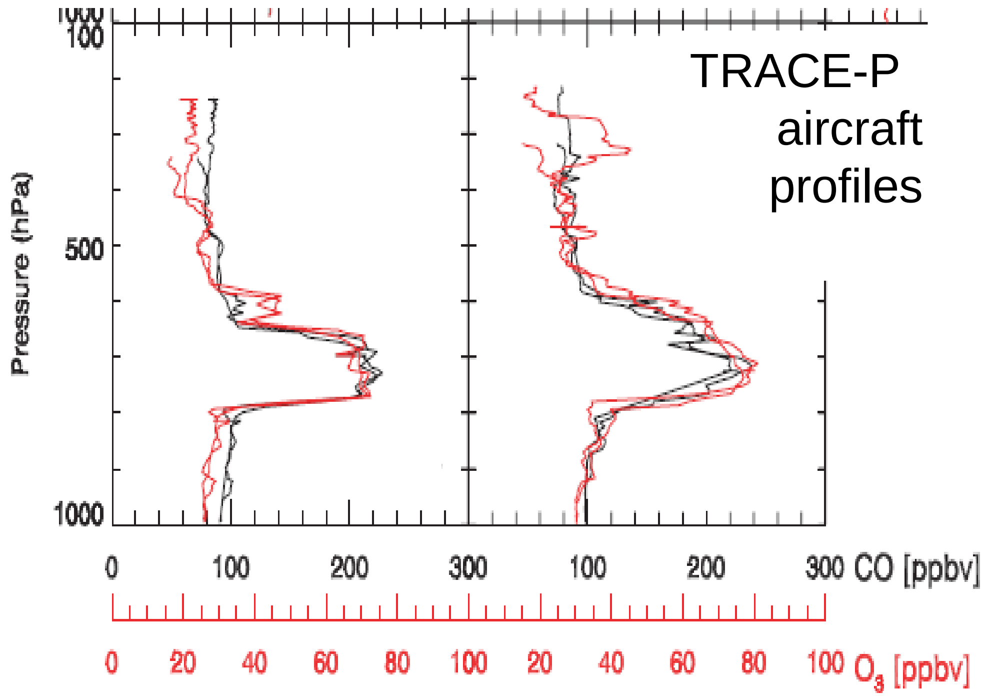
Long-lived chemical plumes in the free troposphere

Free tropospheric CO from AIRS



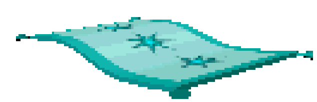


CO and ozone Asian pollution over Pacific



Much of pollution transport on global scale takes place in layers that retain their integrity for over a week, spreading/filamenting horizontally over 1000s of km and vertically over ~1 km

Think of them as “pancakes” or “magic carpets”

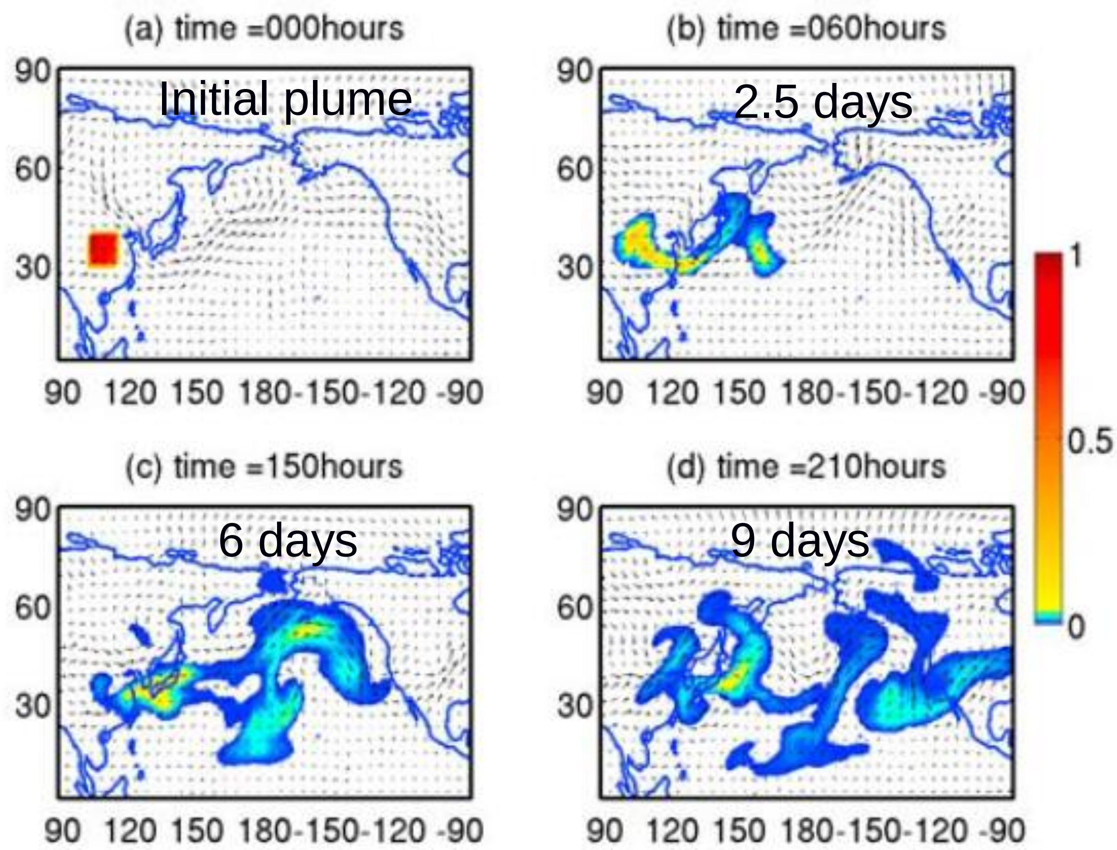


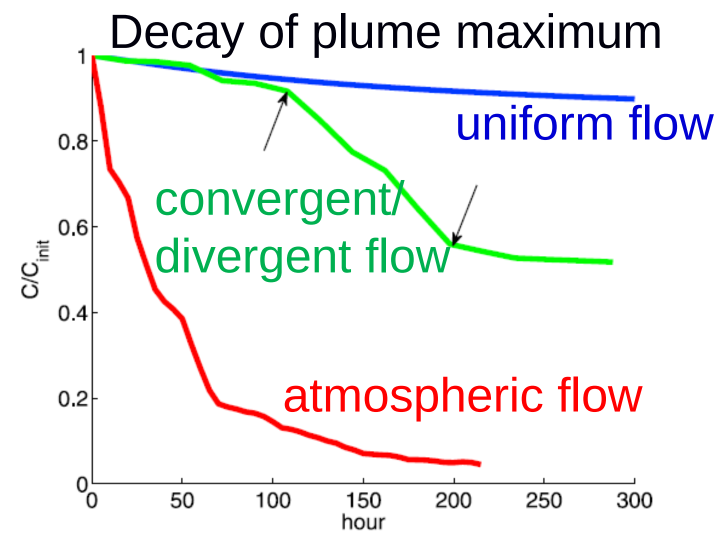
Andreae et al., 1988; Heald et al., 2003

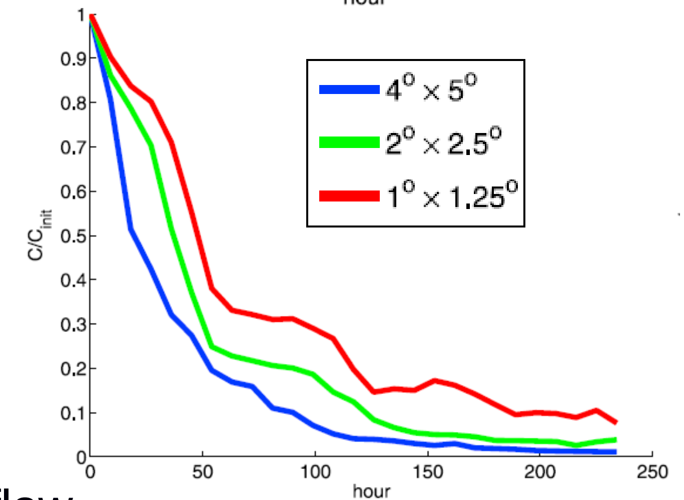
Difficulty of preserving free tropospheric layers in Eulerian models

2-D pure advection $\partial C / \partial t = -\mathbf{u}\nabla C$ of inert Asian plume in GEOS-Chem

Advection scheme is 3rd-order piecewise parabolic method (PPM)



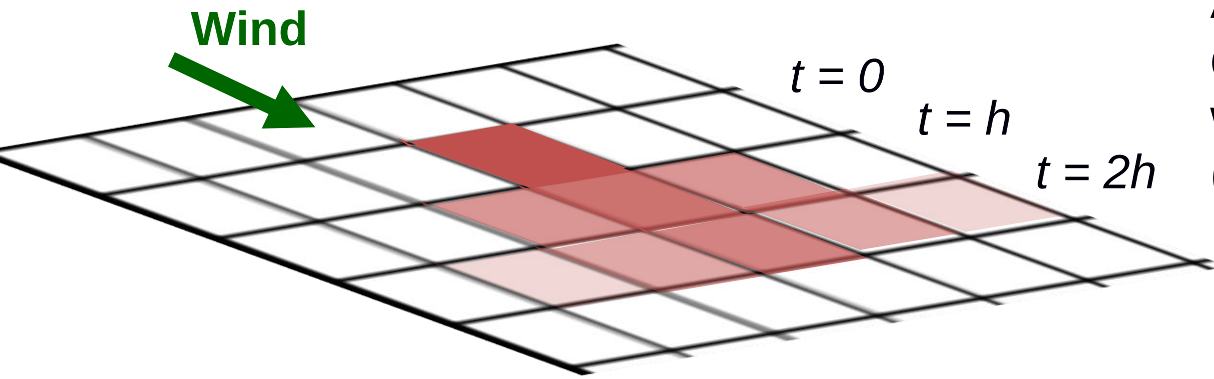




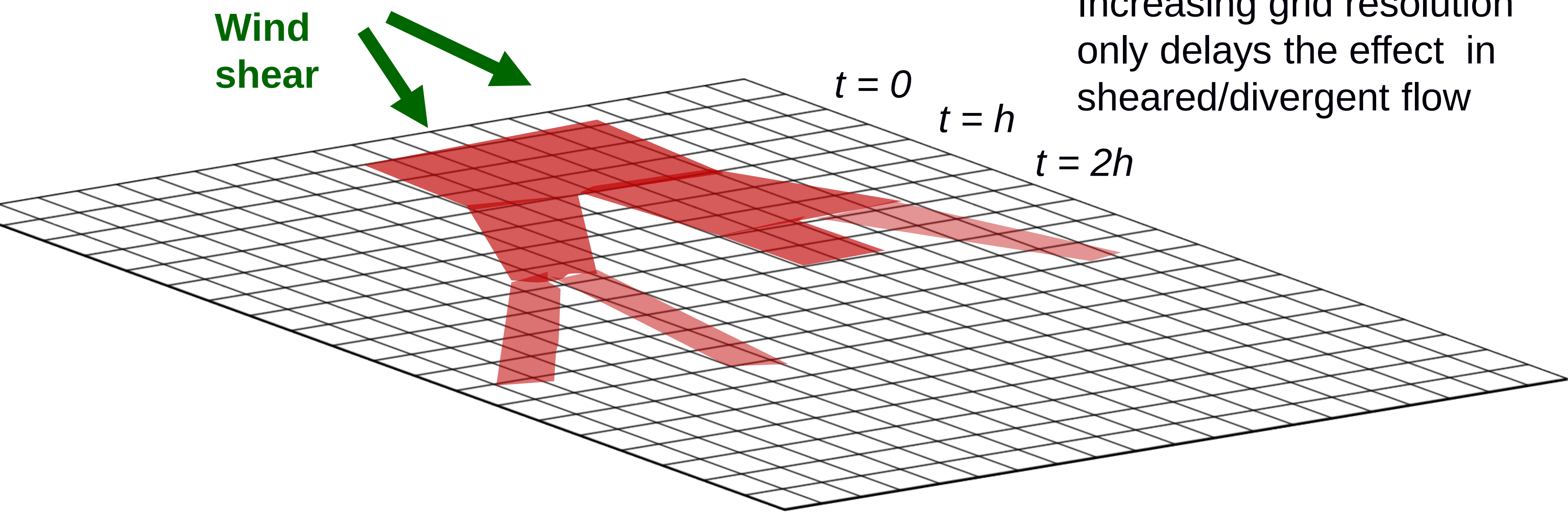
- Advection equation should conserve mixing ratio
- 3rd-order advection scheme fails in divergent/shear flow
- Increasing resolution yields only marginal improvement

Rastigejev et al. [2010]

Why this difficulty? Numerical diffusion as plume shears



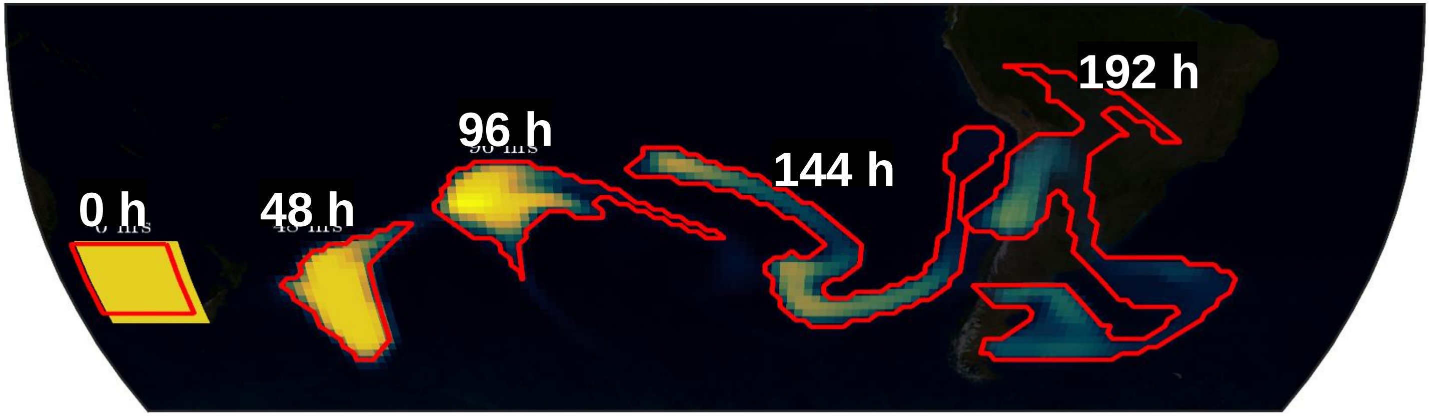
A high-order advection scheme decays to 1st-order when it cannot resolve gradients (plume width ~ grid scale)



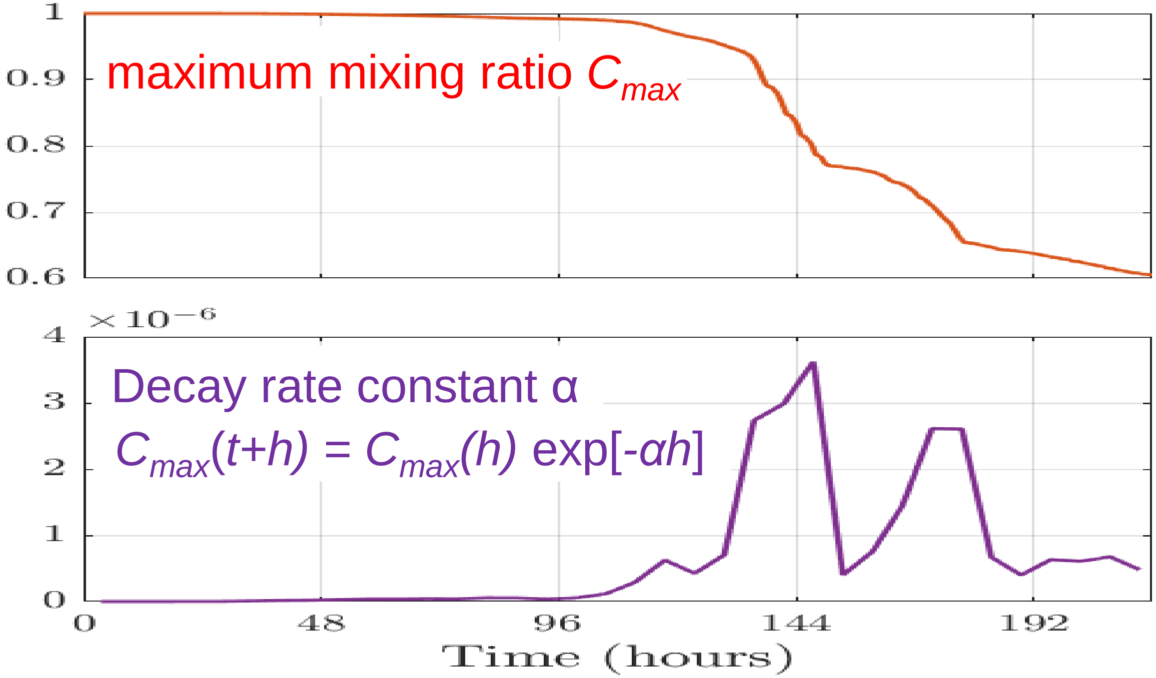
Increasing grid resolution only delays the effect in sheared/divergent flow

Further investigation with 0.25ºx0.3125º version of GEOS-Chem

2-D model grid at 0.25ºx0.3125º, initial plume is 12ºx15º



Color measures volume mixing ratio (VMR)

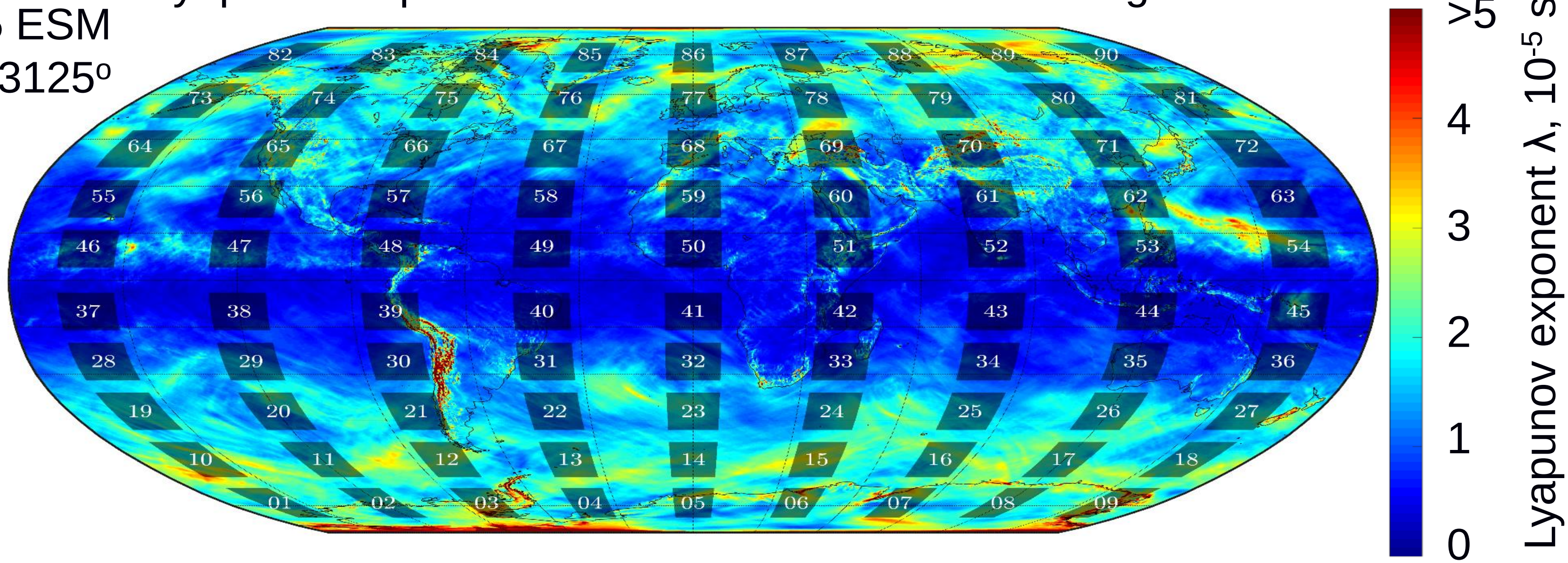


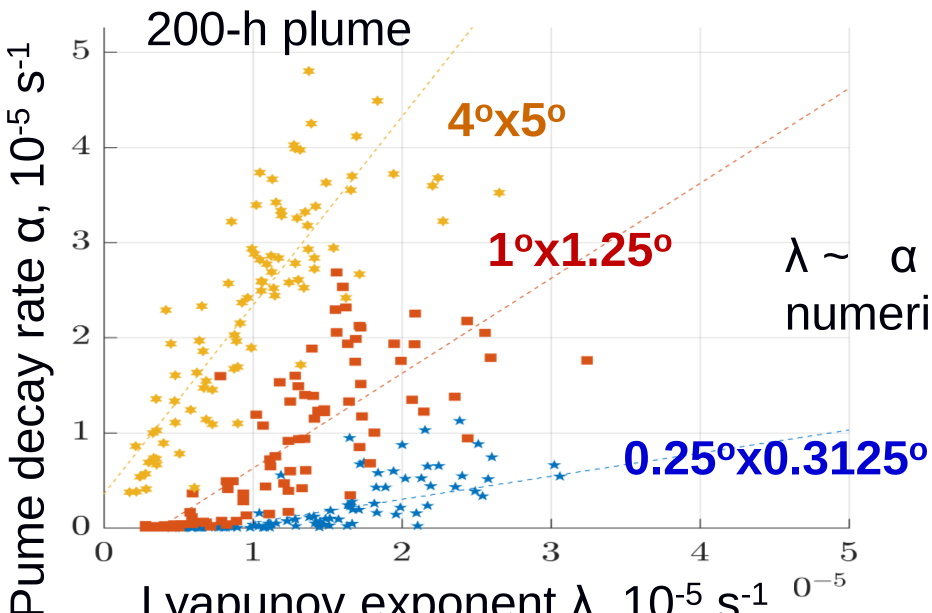
Sebastian Eastham, Harvard

Mapping out the problem with 2-D plumes initialized worldwide

Lyapunov exponents $\lambda = \partial u/\partial x$ measure flow divergence

GEOS-5 ESM
0.25ºx0.3125º



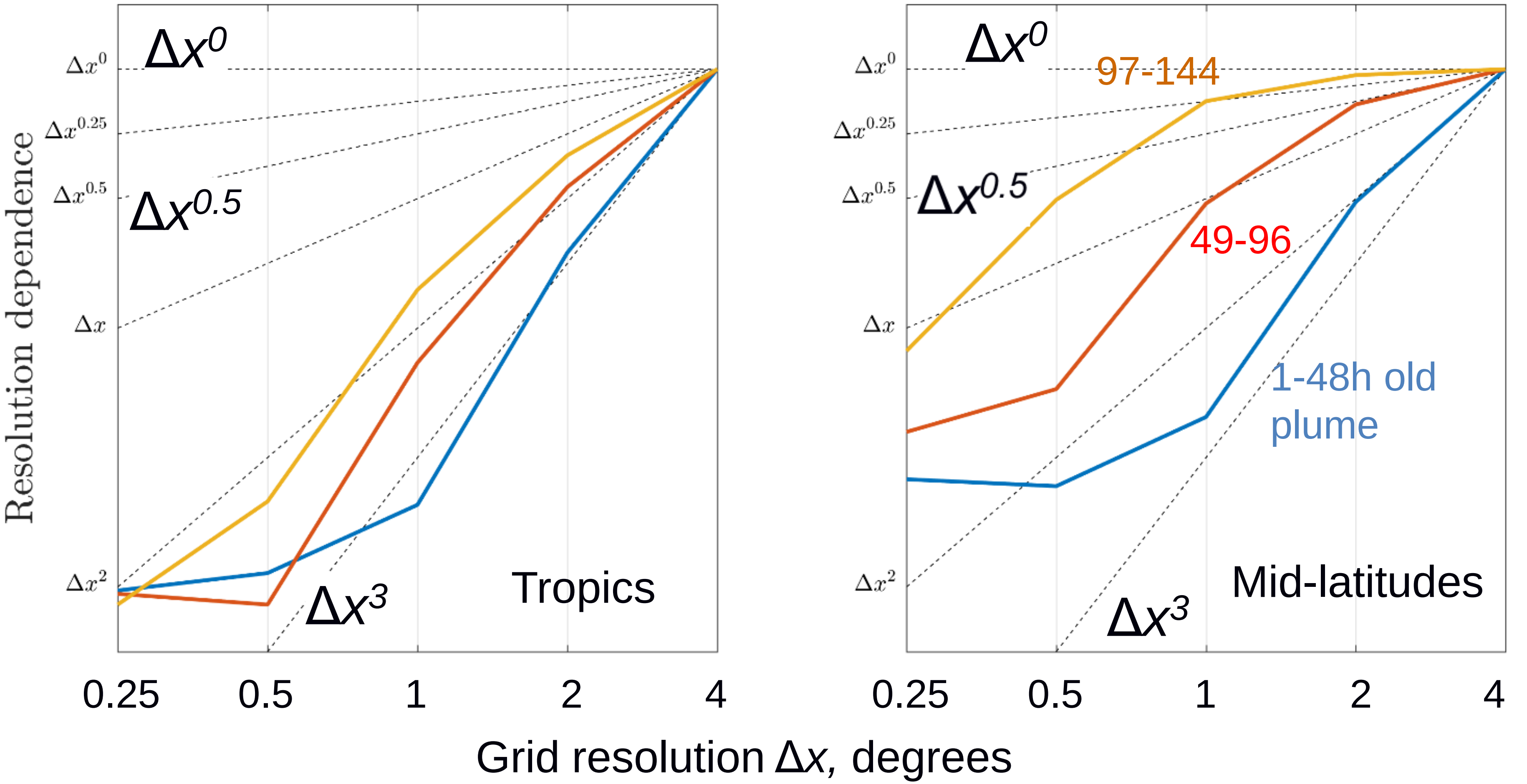


λ ~ α is expected for plumes where numerical diffusion is controlled by stretching

Sebastian Eastham, Harvard

Grid resolution dependence of plume dissipation

How does the plume decay rate constant α depend on the grid resolution Δ*x*?

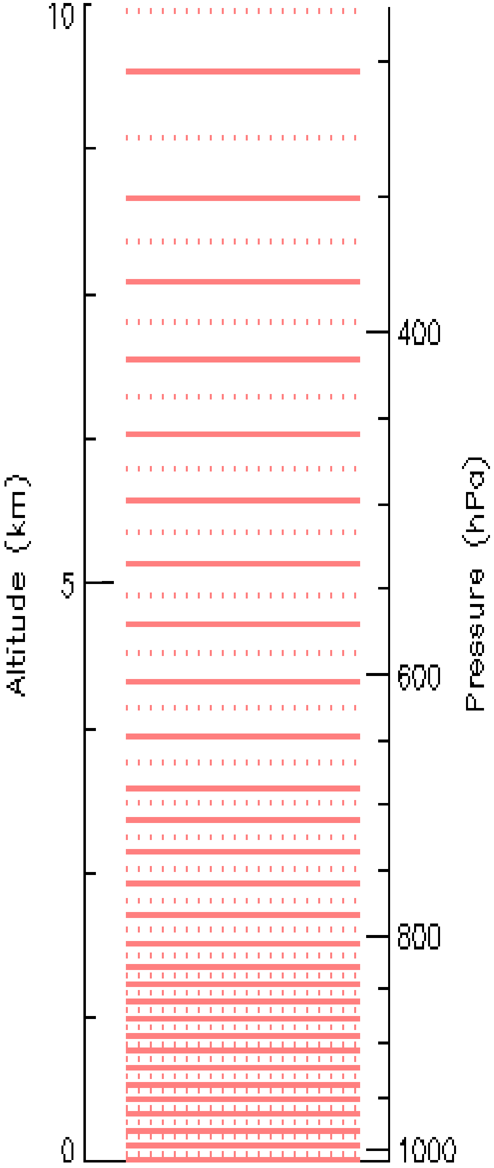


- Numerical diffusion limited by intrinsic numerical accuracy has $\alpha \sim \Delta x^3$
- Numerical diffusion limited by shear/stretching has $\alpha \sim \Delta x^{0.25\text{-}0.5}$

Sebastian Eastham, Harvard

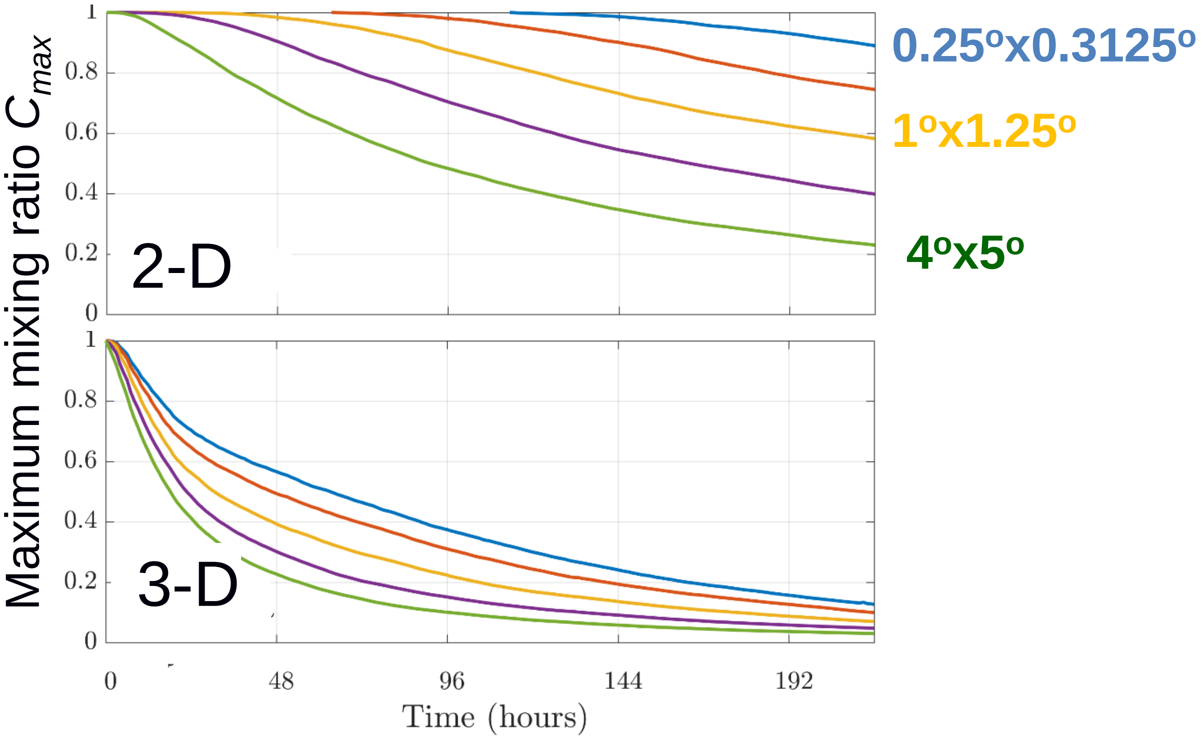
Vertical grid resolution is even more limiting at present

GEOS-5
vertical levels

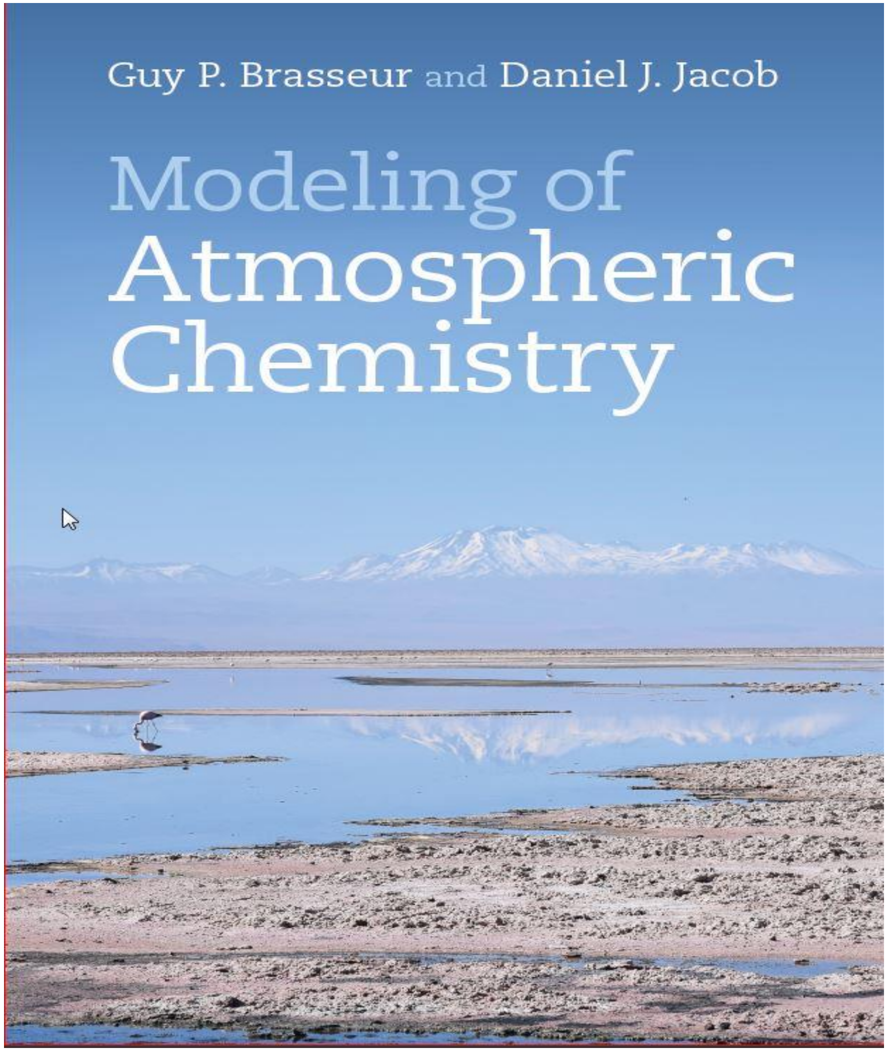


- ESMs prioritize vertical resolution in the boundary layer rather than free troposphere (0.6 km thick in GEOS-5 and ERA-Interim at 4-8 km)
- A typical free tropospheric plume is resolved by only 1-2 vertical layers → large numerical diffusion

Plume decay rate – 3D vs. 2D



Brasseur and Jacob, *Modeling of Atmospheric Chemistry,* Cambridge University Press, 2017



Soon available in all good bookstores!
Email me if you want pre-publication on-line access

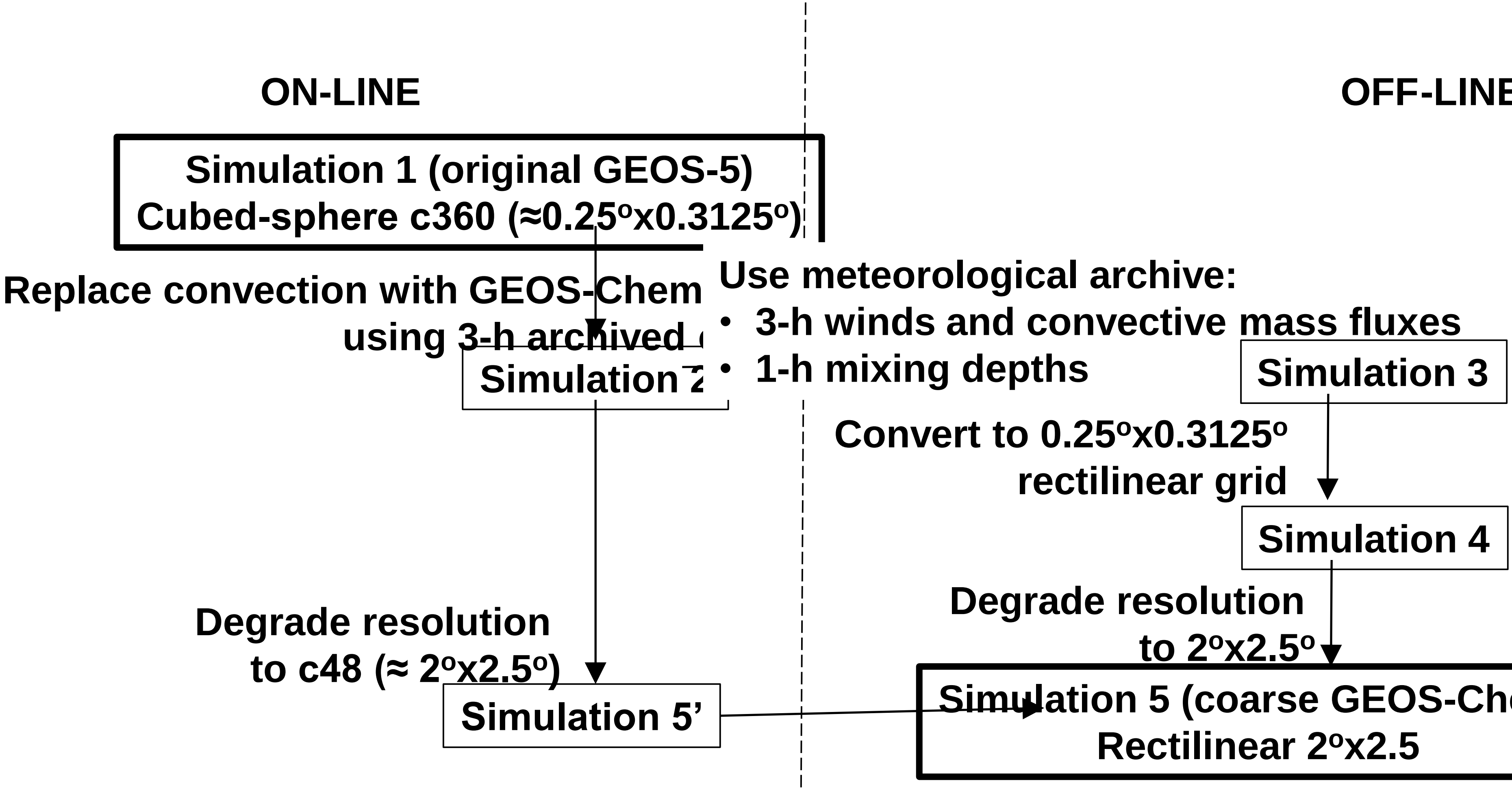
Some take-aways

- Chemical data assimilation has strong clientele for air quality, climate forcing
  - Need to develop new approaches for optimizing surface fluxes
  - Assimilation of aerosol lidar data for mixing depths, CO for winds?

- Chemistry is not that expensive in ESMs
  - It becomes relatively cheaper as model resolution increases
  - It has full scalability in massively parallel architecture
  - It can be done at coarser spatial resolution and time step than dynamics

- Off-line chemical modeling using archived meteorology can be of great value
  - Inverse analyses, sensitivity studies
  - Need to better characterize off-line transport errors as resolution increases

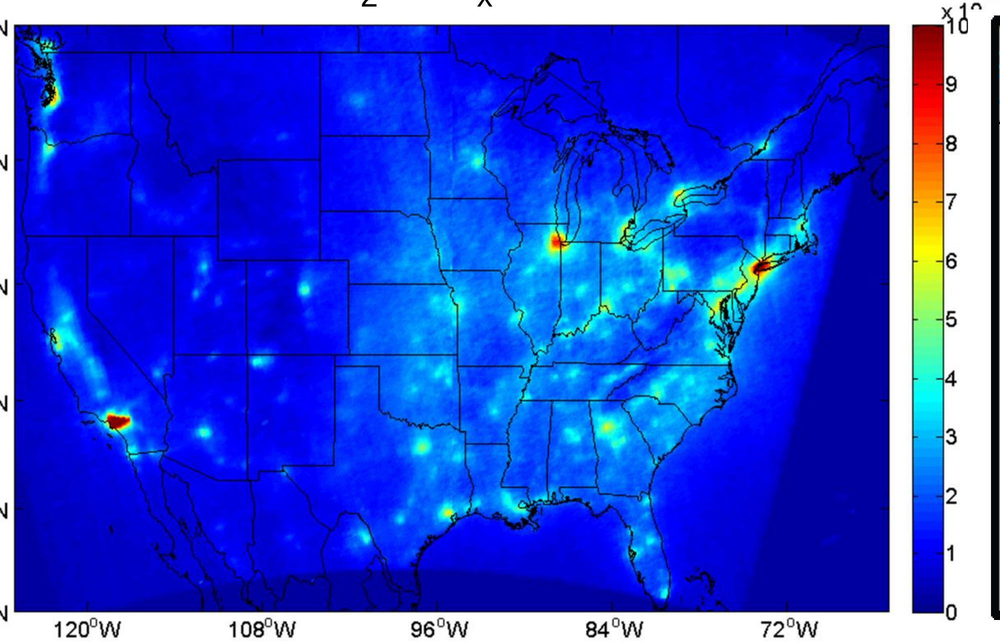
- Transporting intercontinental plumes is a difficult problem for Eulerian models
  - Adding vertical levels to free troposphere is needed



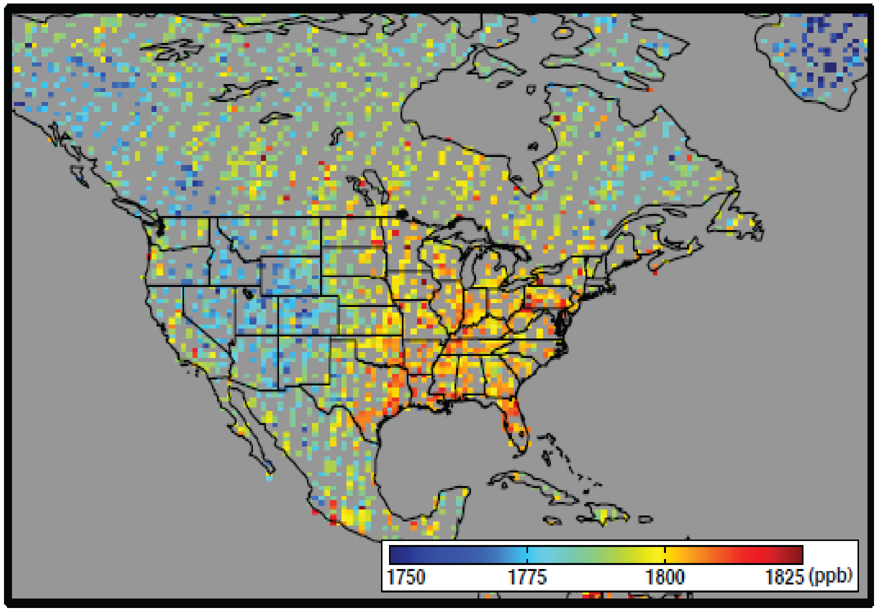
ON-LINE
OFF-LINE
Simulation 1 (original GEOS-5)
Cubed-sphere c360 (≈0.25ºx0.3125º)
Replace convection with GEOS-Chem
using 3-h archived
Simulation 2
Use meteorological archive:
• 3-h winds and convective mass fluxes
• 1-h mixing depths
Simulation 3
Convert to 0.25ºx0.3125º
rectilinear grid
Simulation 4
Degrade resolution
to c48 (≈ 2ºx2.5º)
Simulation 5'
Degrade resolution
to 2ºx2.5º
Simulation 5 (coarse GEOS-Che
Rectilinear 2ºx2.5

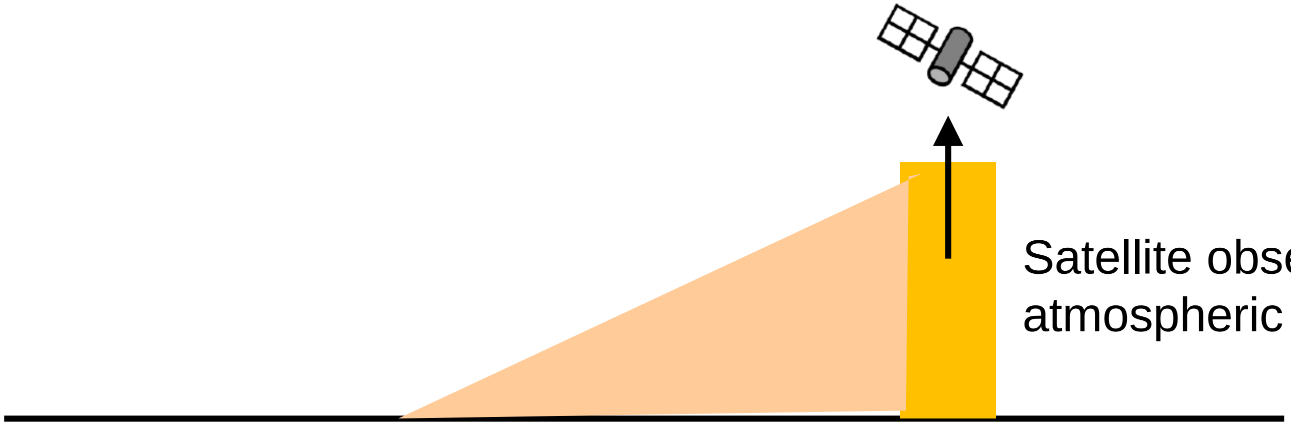
Monitoring emissions from satellite data is emerging priority

OMI NO_2: NO_x emissions



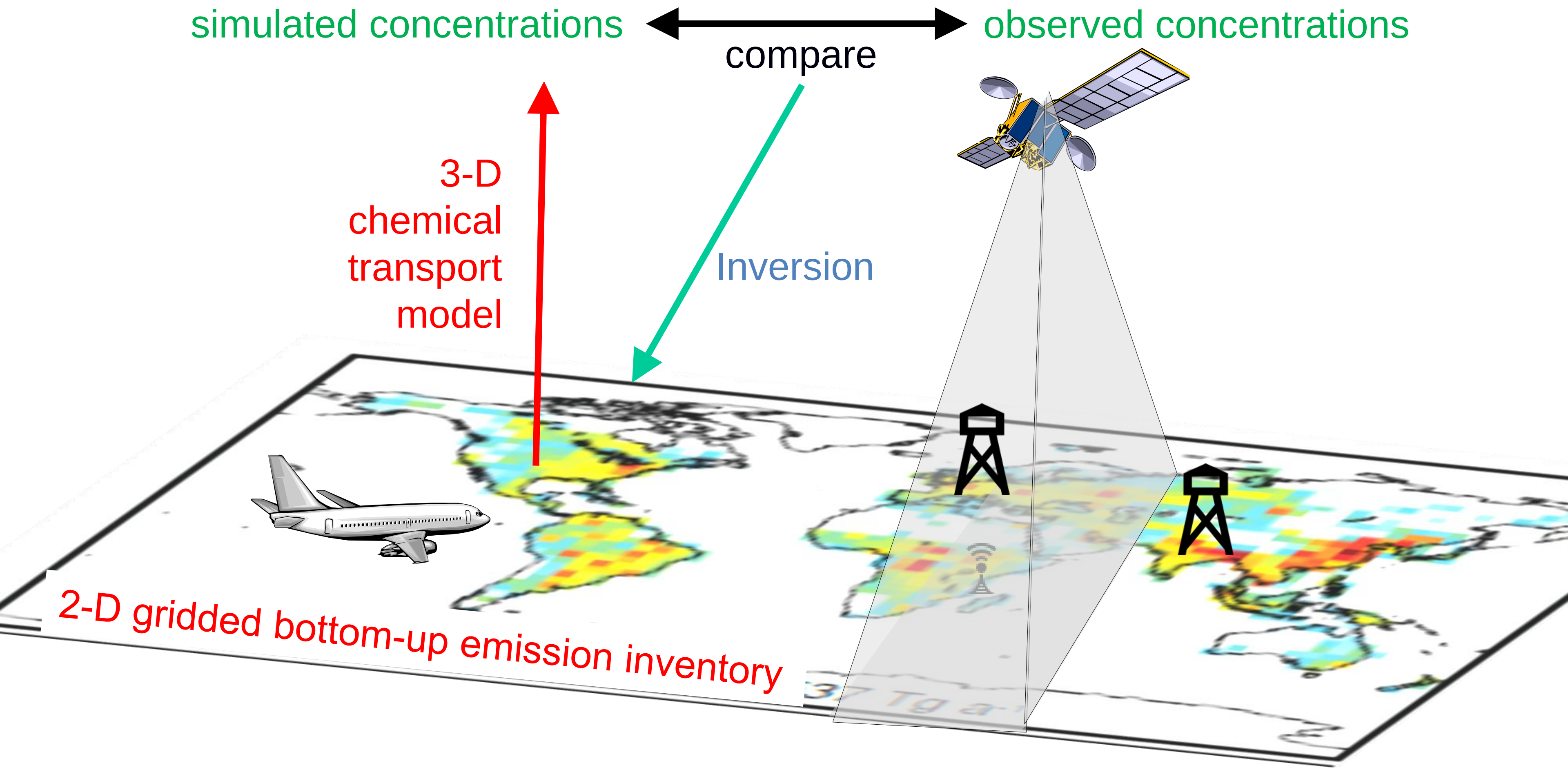
GOSAT methane: methane emissions





Satellite observes atmospheric column

Assimilation of chemical observations to infer emissions



- “Top-down” monitoring of emissions is important for air quality and climate policy
- It is also important for chemical forecasting of air quality
- Near-real-time application allows monitoring of changing emissions

Why are chemical sensitivity studies so important?

Target biases in emissions, chemical parameters

$$\partial C / \partial t = -\mathbf{u}\nabla C + \nabla K \nabla C + \underbrace{P - L + E - D}$$

Biases in:

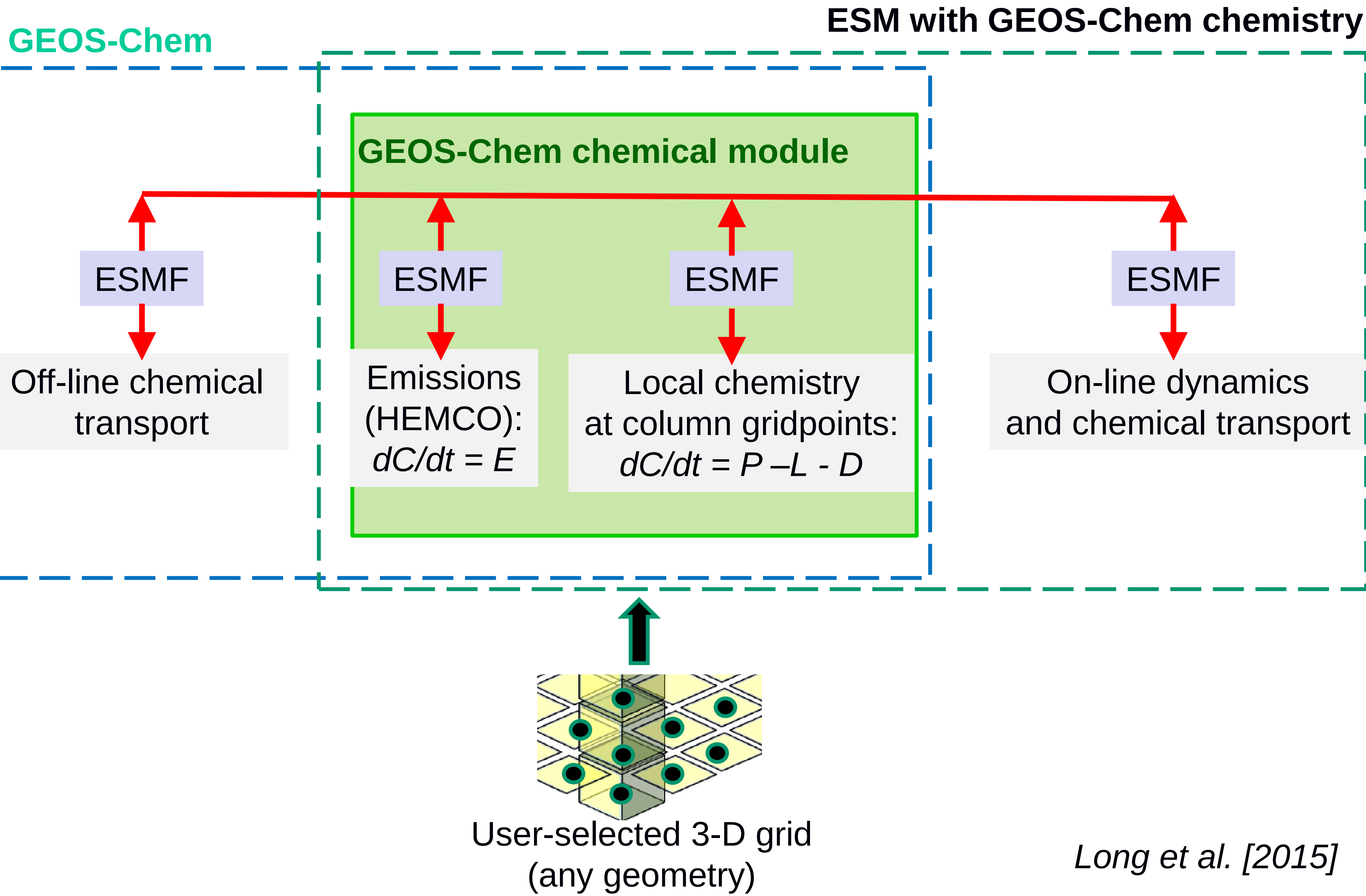
- emissions
- chemical rate constants
- missing/incorrect reactions
- surface uptake
- wet scavenging

Chemical data assimilation ideally requires an unbiased model…

… but chemical errors tend to be systematic

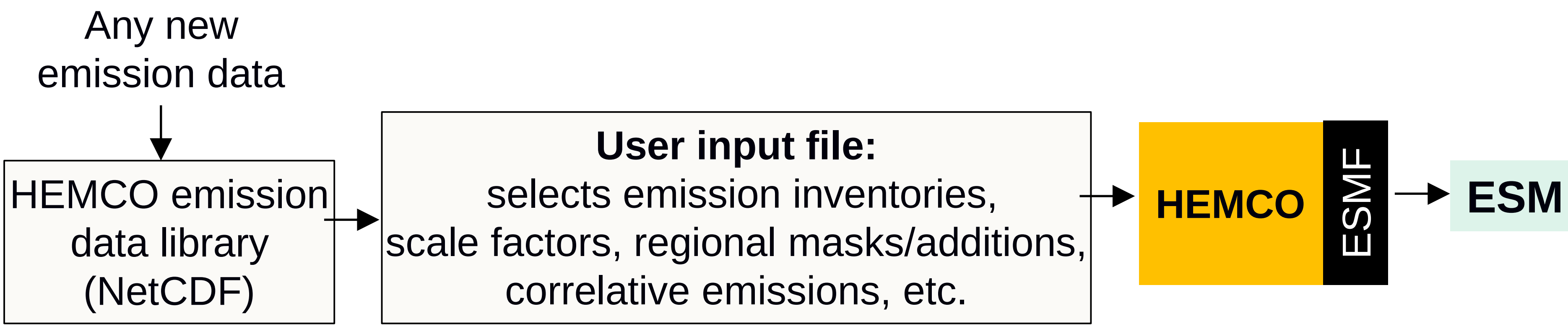
GEOS-Chem as chemical module for Earth System Models:

off-line and on-line simulations use identical code

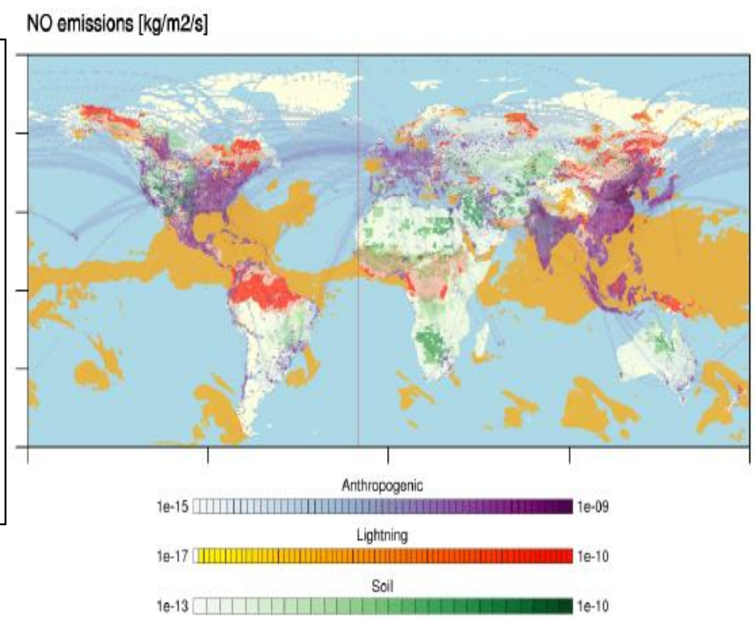


Long et al. [2015]

HEMCO: an ESMF-compliant emission module for Earth System Models



- HEMCO allows to mix/match/scale/overlay any ensemble of emission data for use in ESMs
- Users only edit an input file; no need to edit or compile code, no need to re-grid data
- HEMCO also includes modules for emissions dependent on environmental variables
- HEMCO is now in the GMAO GEOS-5 ESM and is the standard GEOS-Chem emission module

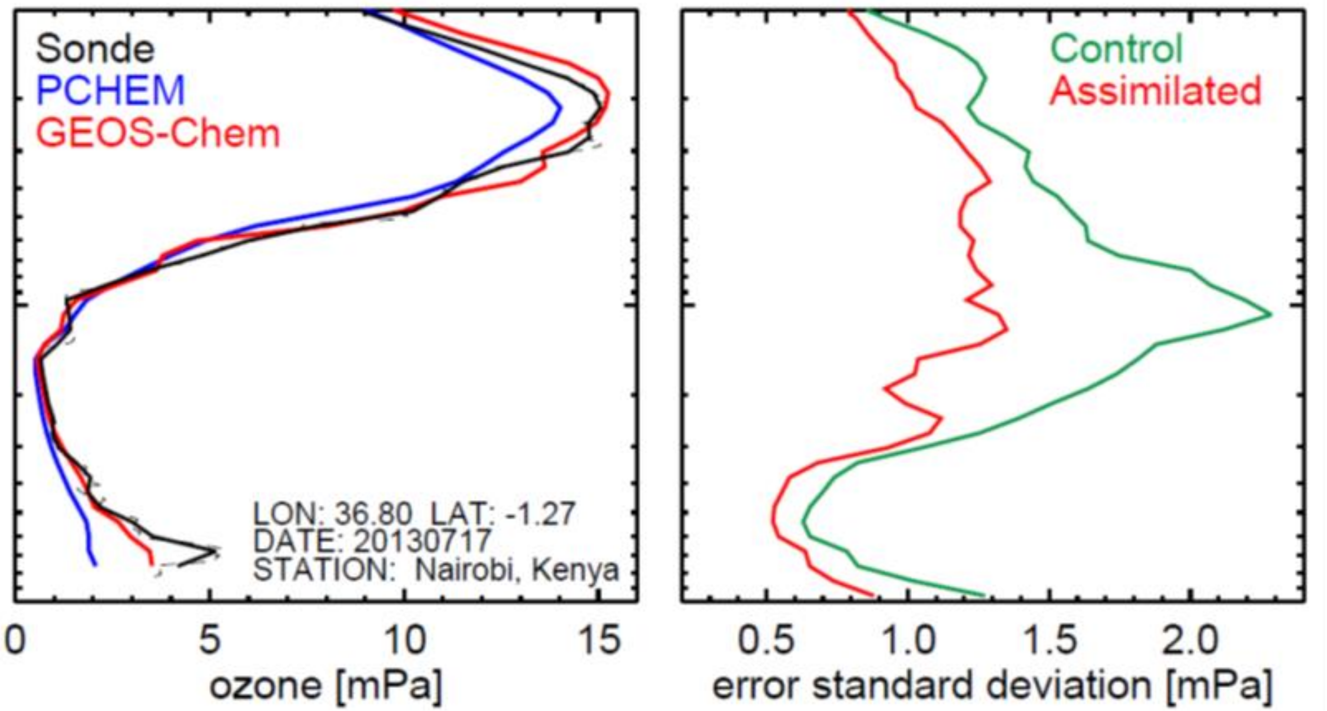


Keller et al. [2014]

Assimilation of OMI+MLS satellite ozone data in the GEOS DAS with GEOS-Chem chemistry module

Comparisons to ozonesondes

Gobal mean error reduction relative to 2013 ozonesondes



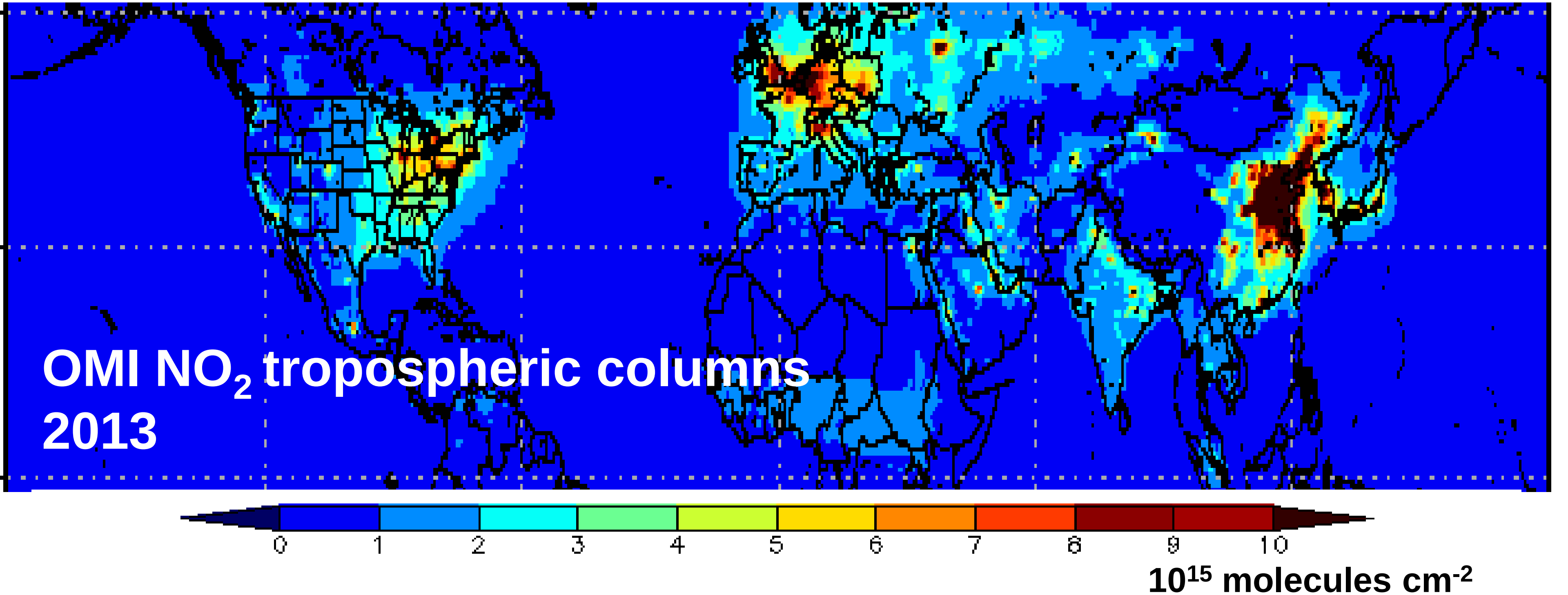
Large model errors in upper troposphere: this is a difficult problem!

- Stratosphere-troposphere exchange
- Lightning NO_x
- Deep convection

Christoph Keller and Kris Wargan, NASA

Emerging era of satellite observations

Tropospheric chemistry has transitioned from data-poor to data-rich over past 15 years



Solar backscatter (column): aerosol, NO_2, ozone, formaldehyde, glyoxal, SO_2, CO, BrO, IO, methane, CO_2

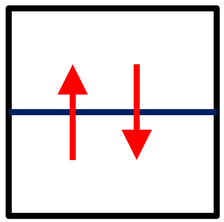
Thermal IR emission: ozone, CO, methane, ammonia,…

Lidar: aerosol. (methane, CO_2, ozone)

Transport errors in off-line models

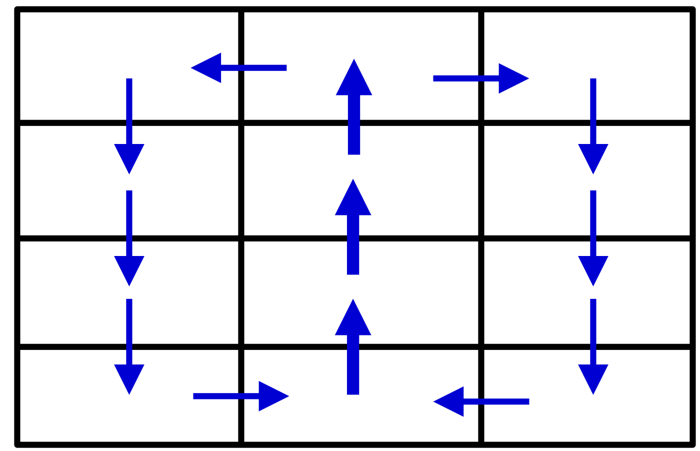
1. Temporal averaging in meteorological archive loses correlation in transient motions:

transient eddies



could be cured by archiving
eddy accumulation

Convective cells resolved by grid-scale advection



not clear how to solve that one

2. Regridding, grid coarsening lead to additional errors

Atmospheric chemistry models solve continuity equations

Eulerian:

$$\partial C / \partial t = -\mathbf{u}\nabla C + \nabla K \nabla C + P - L + E - D$$

change in mxing ratio with time

grid-resolved transport (advection)

subgrid transport (eddies, convection)

chemical production and loss

emission, deposition

Lagrangian:

$$dC / dt = P - L + E - D$$

PROS of Lagrangian over Eulerian models:

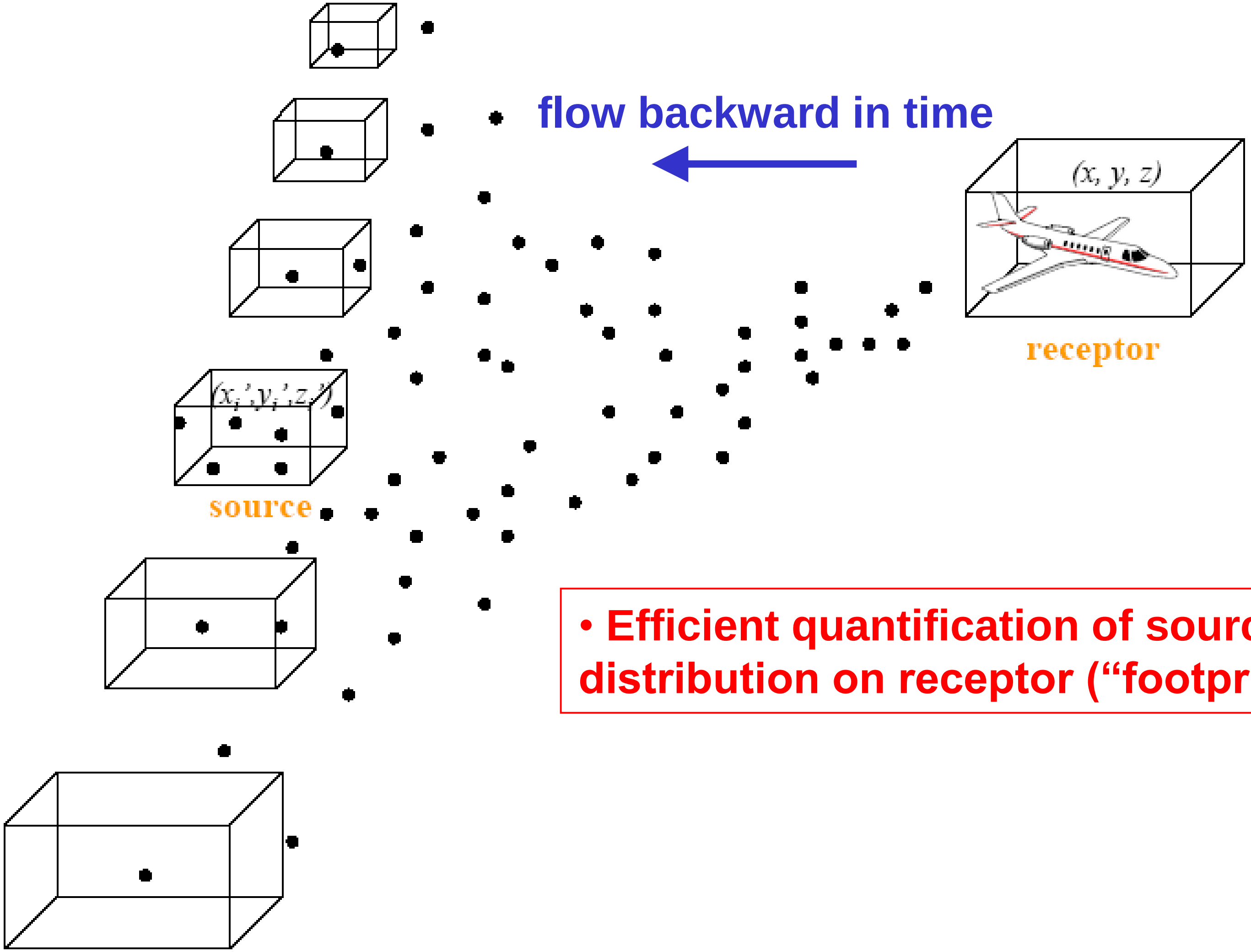
- stable for any wind speed
- no numerical diffusion
- easily track air parcel histories (receptor-oriented problems)
- easy to parallelize

CONS:

- need very large # points for statistics
- inhomogeneous representation of domain
- individual trajectories do not mix
- nonlinear chemistry is problematic
- no on-line coupling with Eulerian meteorological model

Lagrangian receptor-oriented modeling

Run Lagrangian model backward from receptor location, with points released at receptor location only



- **Efficient quantification of source influence distribution on receptor ("footprint")**

Atmospheric chemistry models solve continuity equations

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change in mxing ratio with time | grid-resolved transport (advection) | subgrid transport (eddies, convection) | chemical production and loss | emission, deposition

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