EPS200: Atmospheric Chemistry and Physics

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Chemical transport models

Atmospheric chemistry models

- **Preparatory reading: chapter 3 of *Intro to Atmospheric Chemistry***
- **Further reading (optional!):**
  - **Chapters 1 (continuity equation) and chapter 2 (transport operator) of *Chemical Transport Models***
  - **Chapter 4 (model equations and numerical approaches) of *Mathematical Modeling of Atmospheric Chemistry***

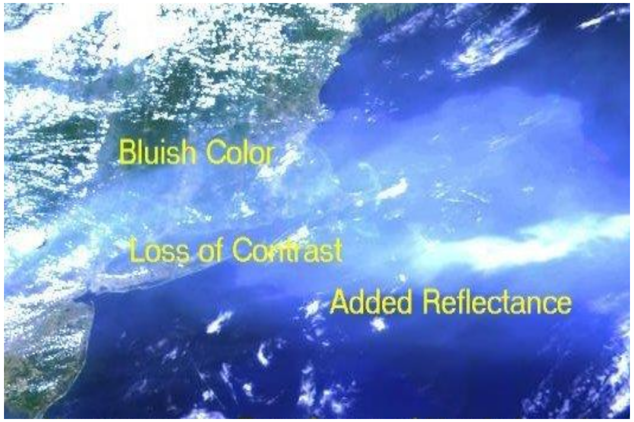
Available at http://acmg.seas.harvard.edu/education

Special case of atmospheric aerosols

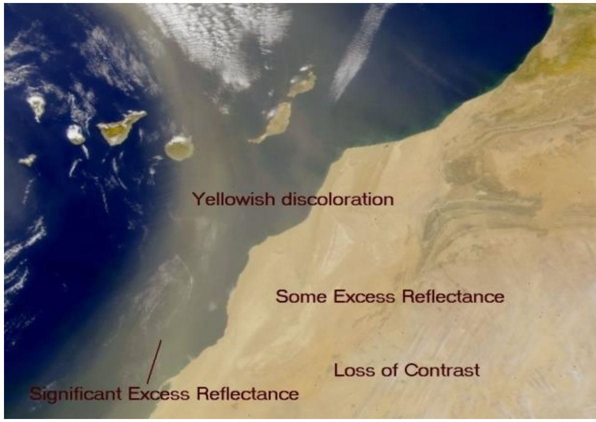
The atmosphere contains suspended solid and liquid particles (*aerosols)* in addition to gases; these particles are the "visible" components of the atmosphere

California fire plumes

Pollution off U.S. east coast



Dust off West Africa



What about clouds? Clouds are made up of water droplets or ice crystals (1-100 μm), much larger than typical aerosols (0.01-10 μm). They are technically aerosols but have unique properties and are in practice considered separately.

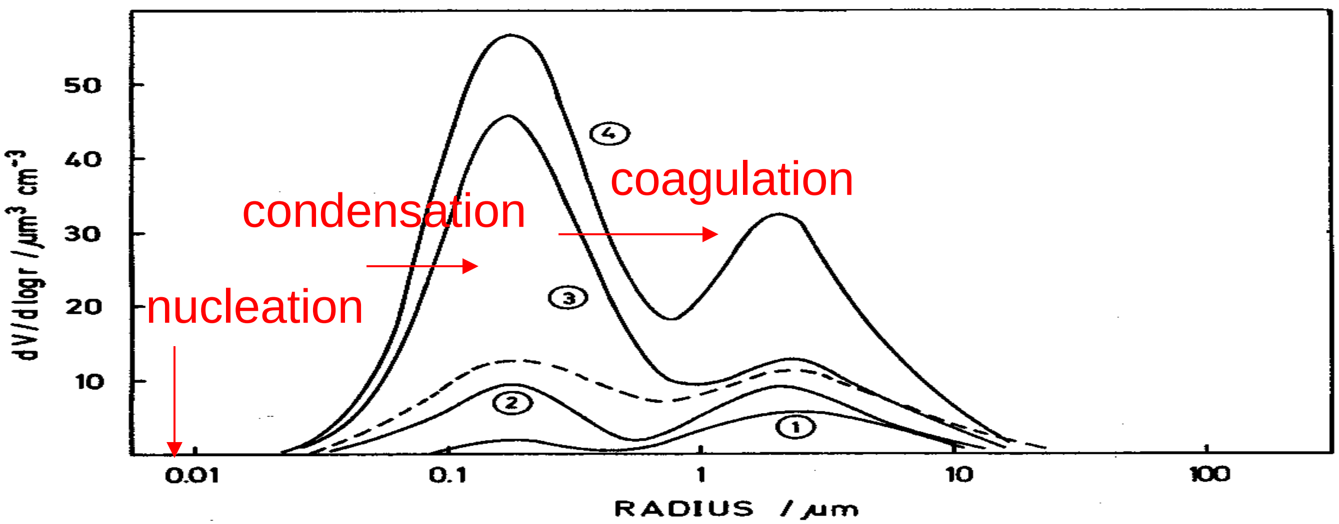
Applying the continuity equation to aerosols

• A given aerosol particle is characterized by its size, shape, phases, and chemical composition – large number of variables!

• Measures of aerosol concentrations must be given in some integral form, by summing over all particles present in a given air volume that have a certain property

• If evolution of the size distribution is not resolved, continuity equation for aerosol species can be applied in same way as for gases

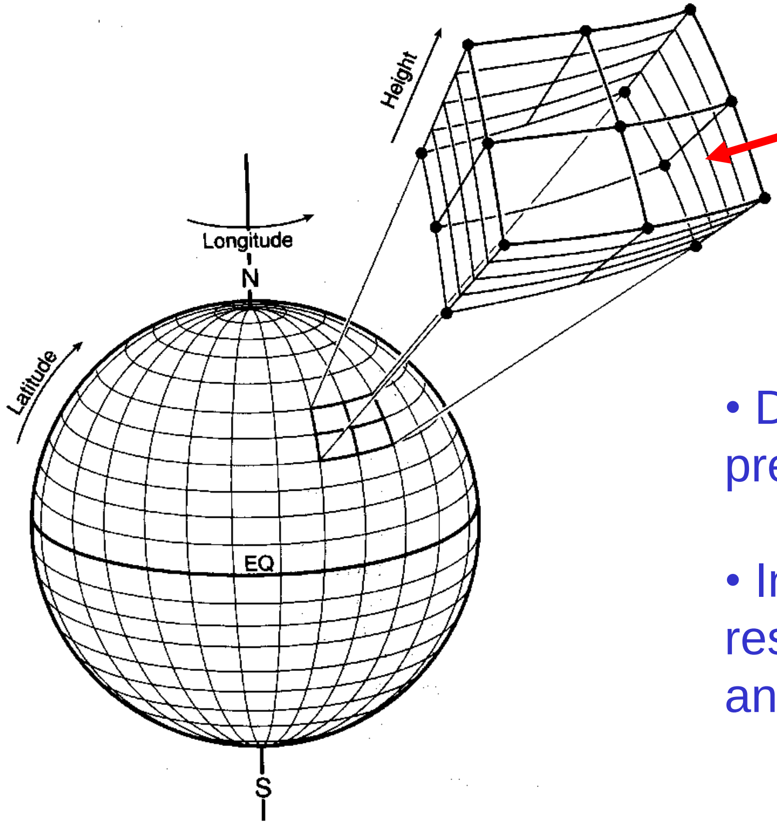
• Simulating the evolution of the aerosol size distribution requires inclusion of nucleation/growth/coagulation terms in P_i and L_i, and size characterization either through size bins or moments.



Typical aerosol size distributions by volume

EULERIAN MODELS PARTITION ATMOSPHERIC DOMAIN INTO GRIDBOXES

This discretizes the continuity equation in space



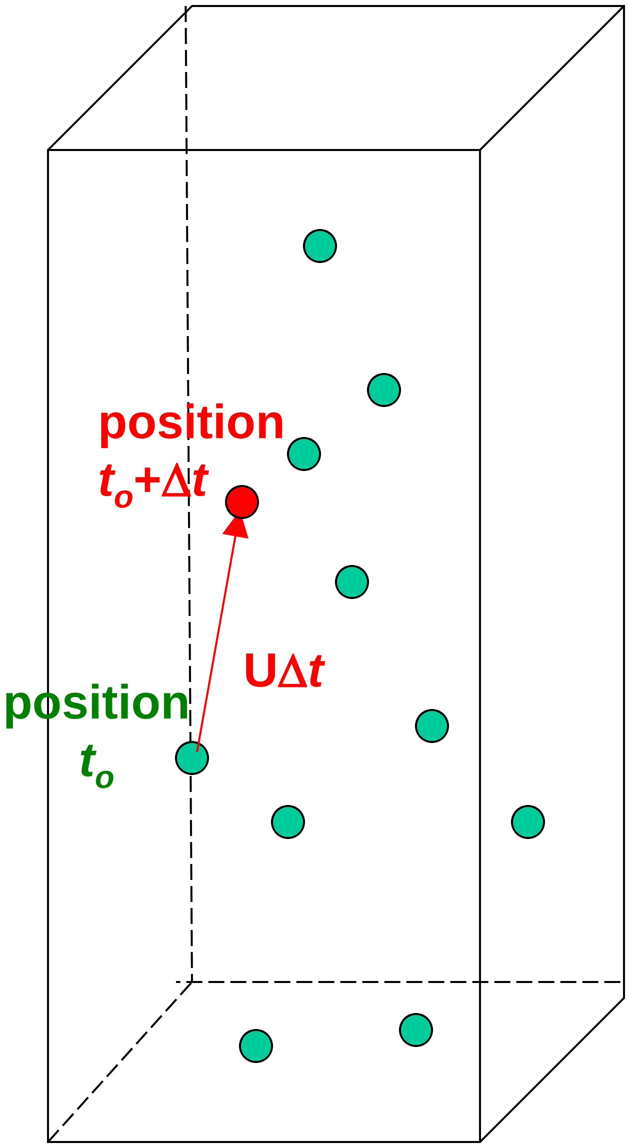
Solve continuity equation for individual gridboxes

- Detailed chemical/aerosol models can presently afford -10^6 gridboxes

- In global models, this implies a horizontal resolution of ~ 1^o (~100 km) in horizontal and ~ 1 km in vertical

- Chemical Transport Models (CTMs) use external meteorological data as input
- General Circulation Models (GCMs) compute their own meteorological fields

LAGRANGIAN MODELS TRACK TRANSPORT OF POINTS IN MODEL DOMAIN (NO GRID)



- Transport large number of points with trajectories from input meteorological data base (**U**) over time steps Δt
- Points have mass but no volume
- Determine local concentrations as the number of points within a given volume
- Nonlinear chemistry requires Eulerian mapping at every time step (semi-Lagrangian)

PROS over Eulerian models:

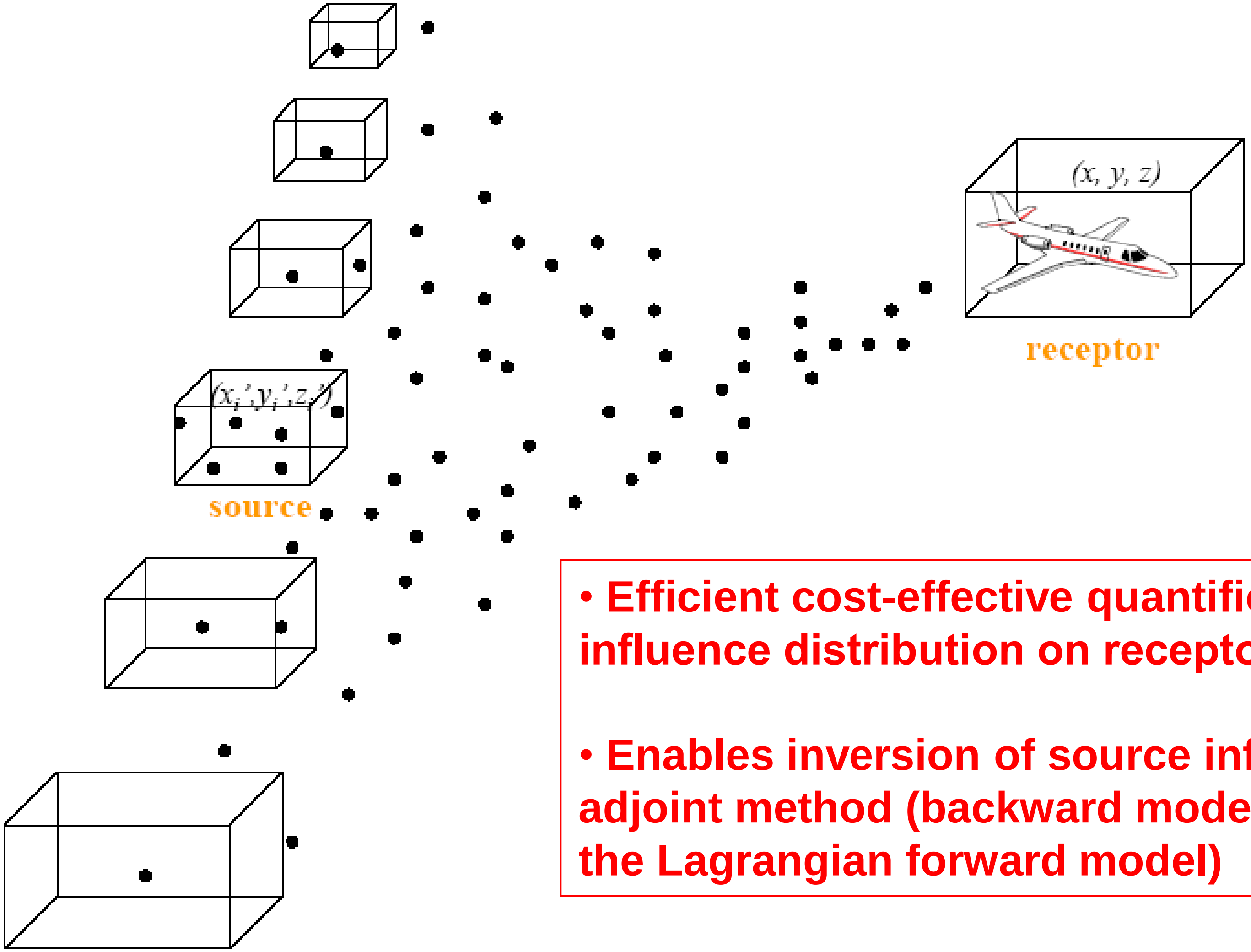
- no Courant number restrictions
- no numerical diffusion/dispersion
- easily track air parcel histories
- invertible with respect to time

CONS:

- need very large # points for statistics
- inhomogeneous representation of domain
- convection is poorly represented
- nonlinear chemistry is problematic

LAGRANGIAN RECEPTOR-ORIENTED MODELING

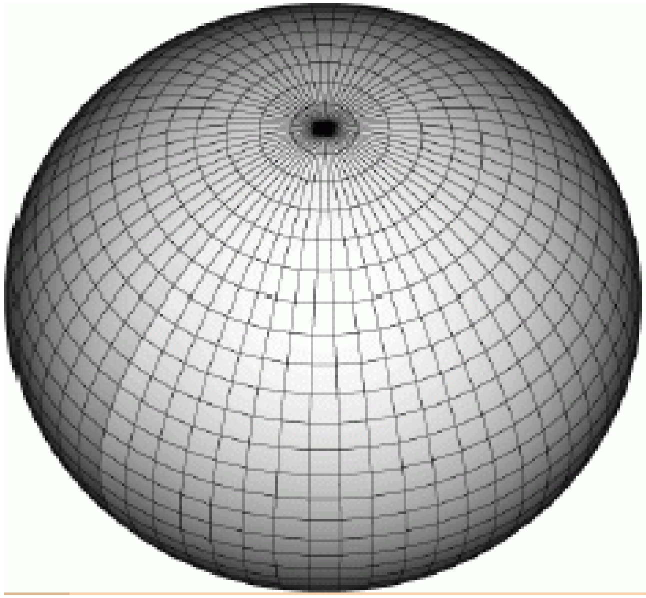
Run Lagrangian model backward from receptor location, with points released at receptor location only

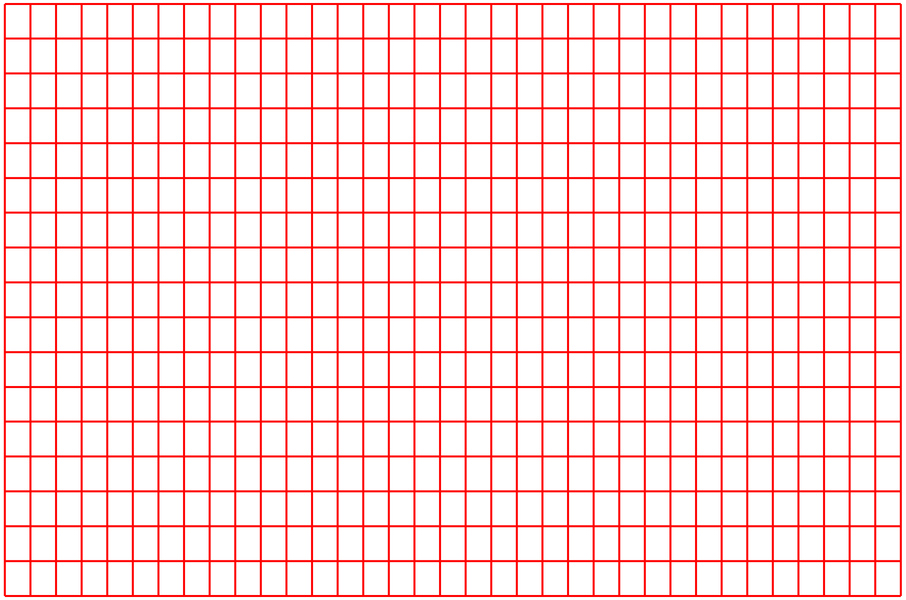


- **Efficient cost-effective quantification of source influence distribution on receptor ("footprint")**

- **Enables inversion of source influences by the adjoint method (backward model is the adjoint of the Lagrangian forward model)**

Global horizontal grids for Eulerian models

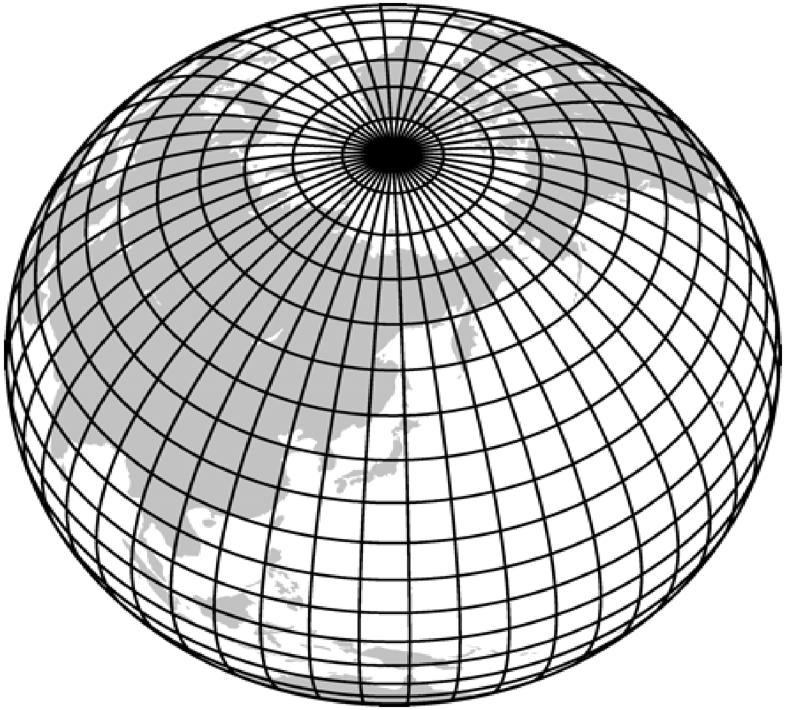




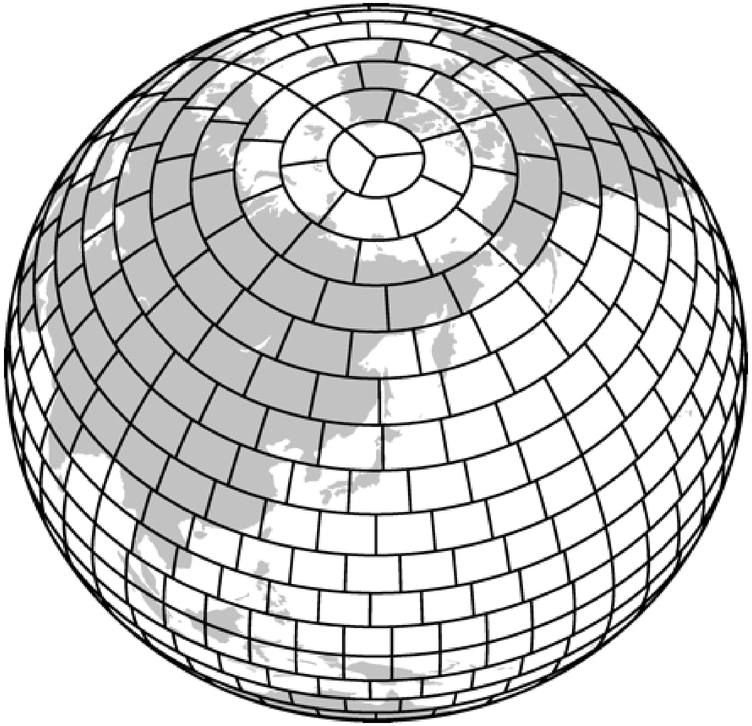
Latitude-Longitude grid: spherical representation (left) and plane projection (right)

Pro: *x* and *y* axes are orthogonal
Con: singularities at the poles

Reduced grid to avoid polar singularities



Regular lat-lon grid



Reduced lat-lon grid: grid spacing increases with increasing latitude

Washington et al. [2009]

Global horizontal grids for Eulerian models (cont.)

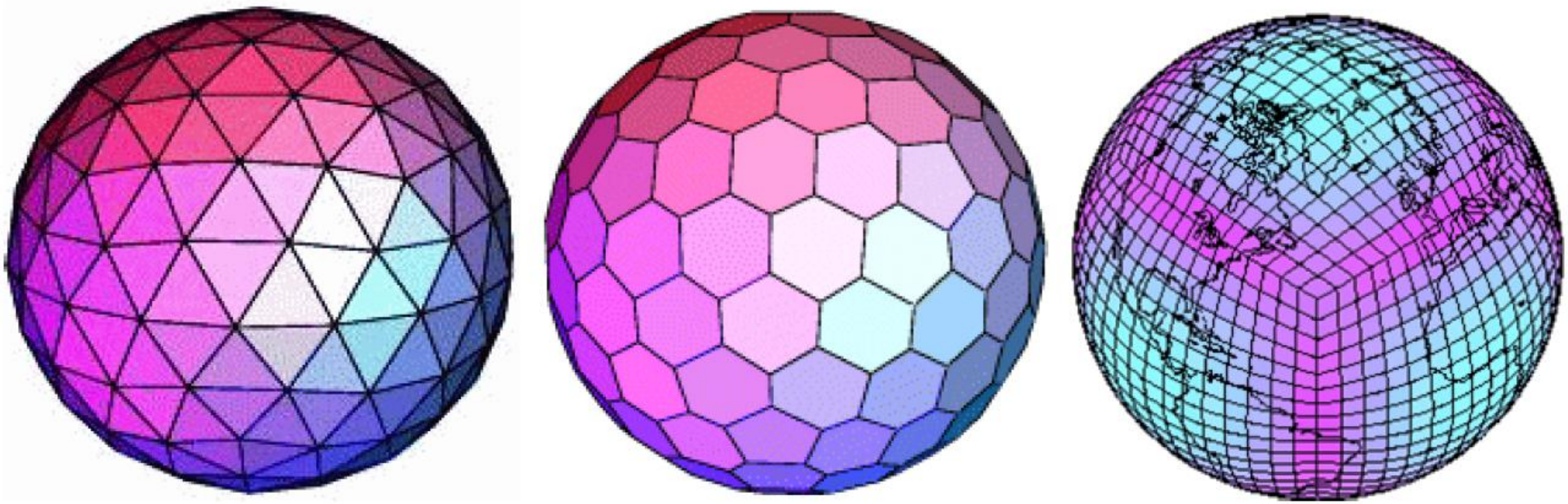
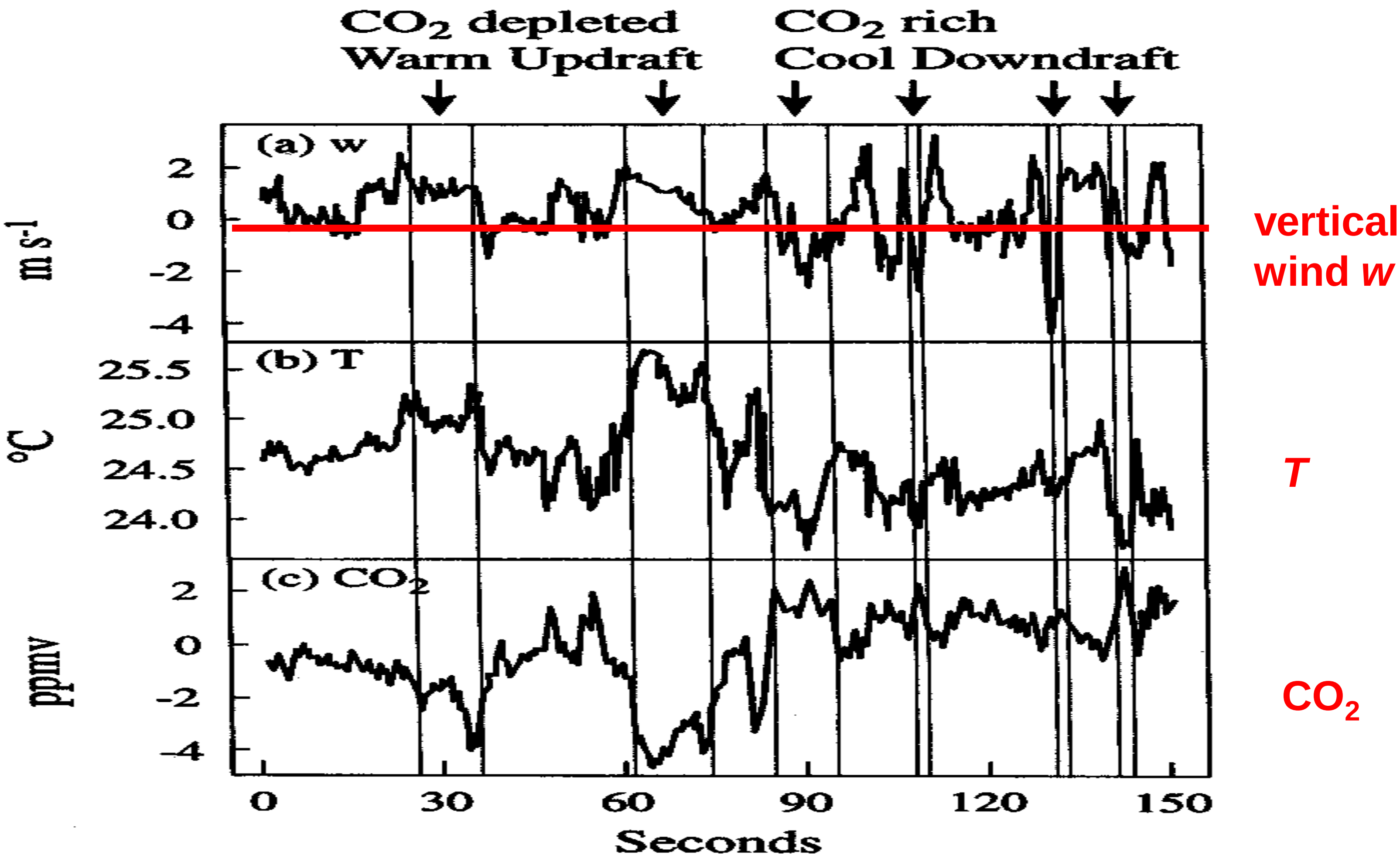


Figure 4.15. Examples of grids: icosahedral triangular (left), icosahedral hexagonal (middle) and cubed sphere (right).

Summer daytime CO_2 flux observations at Harvard Forest, Massachusetts



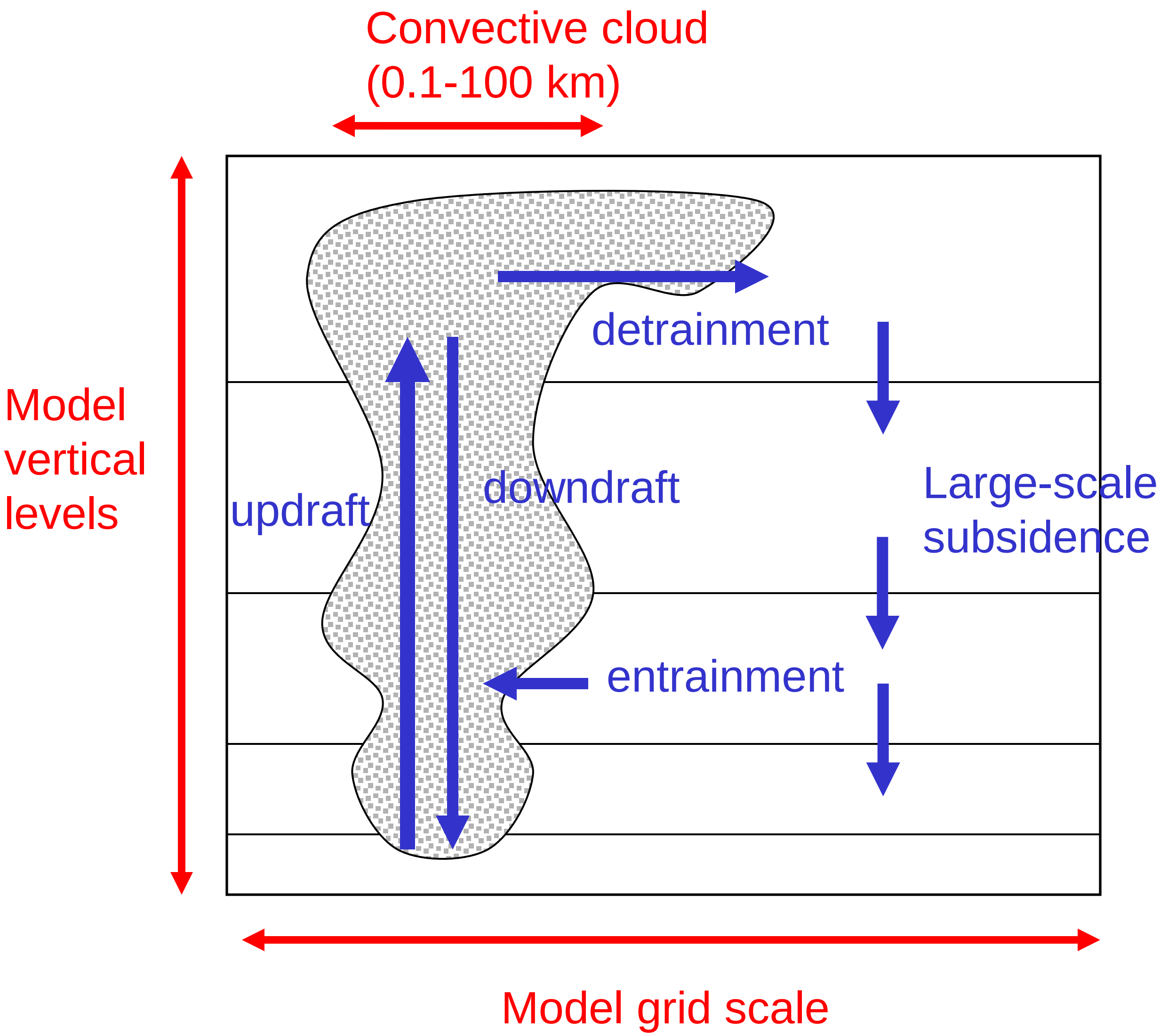
Gaussian dispersion of pollution plumes

California fire plumes, Oct 2004

Industrial plumes

DEEP CONVECTION

- Organized but subgrid (except in cloud-resolving model with ~1 km resolution)
- Requires non-local parameterization

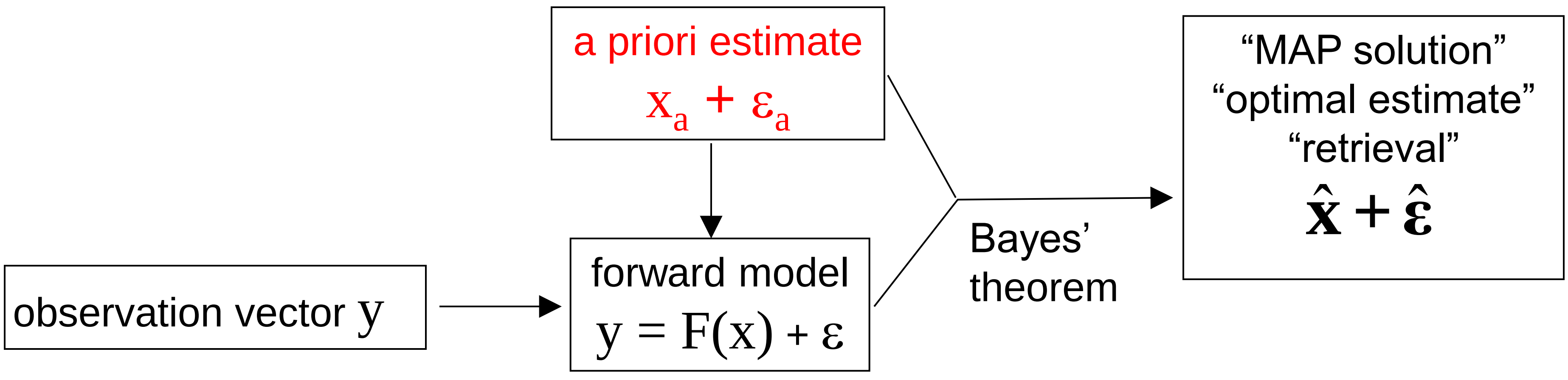


Questions

1. The Eulerian form of the continuity equation is a first-order PDE in four dimensions. What are suitable boundary conditions for each of these dimensions in a global model?
2. A Lagrangian air parcel contains two gas-phase species A and B. Can we calculate the rate of reaction A + B -> products in that air parcel?
3. A plane flies a series of stacked horizontal legs in the boundary layer measuring the CO_2 vertical flux as *wn* where *w* is the vertical velocity and *n* is the number density of CO_2. As long as the plane stays in the boundary layer, do you expect the vertical flux to stay constant with altitude?
4. Pollution plumes dilute much more quickly in a stretched (acccelerating) flow than in a uniform flow. Why?
5. A convective updraft has a base at 20^{o}C and top at -40^{o}C. Assuming no entrainment in the updraft, what is the scavenging efficiency of water in the updraft?

THE INVERSE MODELING PROBLEM

Optimize values of an ensemble of variables (*state vector* x) using observations:



THREE MAIN APPLICATIONS FOR ATMOSPHERIC COMPOSITION:

1. Retrieve atmospheric concentrations (x) from observed atmospheric radiances (y) using a radiative transfer model as forward model
2. Invert sources (x) from observed atmospheric concentrations (y) using a CTM as forward model
3. Construct a continuous field of concentrations (x) by assimilation of sparse observations (y) using a forecast model (initial-value CTM) as forward model

BAYES' THEOREM: FOUNDATION FOR INVERSE MODELS

$P(\mathrm{x})$ = probability distribution function (pdf) of x

$P(\mathrm{x,y})$ = pdf of (x,y)

$P(\mathrm{y|x})$ = pdf of y given x

$$P(\mathrm{x,y})d\mathrm{x}d\mathrm{y} = P(\mathbf{x})d\mathbf{x}P(\mathbf{y}\,|\,\mathbf{x})d\mathbf{y}$$

$$P(\mathrm{x,y})d\mathrm{x}d\mathrm{y} = P(\mathbf{y})d\mathbf{y}P(\mathbf{x}\,|\,\mathbf{y})d\mathbf{x}$$

$$\Rightarrow \quad \underbrace{P(\mathbf{x}\,|\,\mathbf{y})}_{a\ posteriori\ \text{pdf}} = \frac{\overbrace{P(\mathbf{y}\,|\,\mathbf{x})}^{\text{observation } pdf}\overbrace{P(\mathbf{x})}^{a\ priori\ \text{pdf}}}{\underbrace{P(\mathbf{y})}_{\text{normalizing factor (unimportant)}}}$$

Bayes' theorem

Maximum *a posteriori* (MAP) solution for x given y is defined by $\max(P(\mathbf{x}\,|\,\mathbf{y}))$

⇨ solve for $\nabla_{\mathbf{x}} P(\mathbf{x}\,|\,\mathbf{y}) = \mathbf{0}$