

Lecture 13. Mathematical Models

1. Model types.

Mathematical models provide the necessary framework for integration of our understanding of individual atmospheric processes and study of their interactions.

Note that atmosphere is a complex reactive system in which numerous physical and chemical processes occur simultaneously.

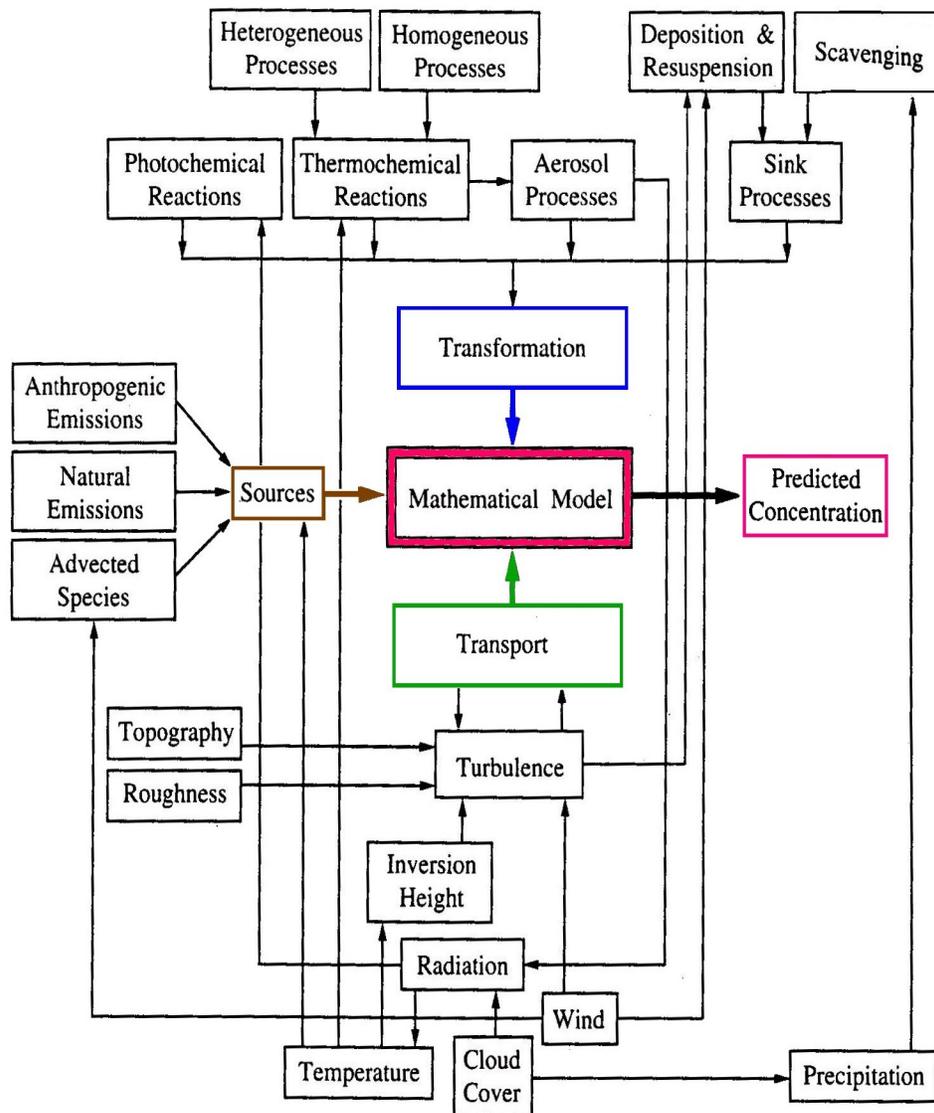


Figure 1 Elements of mathematical atmospheric model (Seinfeld & Pandis, 1998).

Model scales

- Atmospheric chemical transport models are defined according to their spatial scale:

Model	Typical domain scale	Typical resolution
Microscale	200x200x100 m	10 m
Mesoscale(urban)	100x100x5 km	1 km
Regional	1000x1000x10 km	10 km
Synoptic(continental)	3000x3000x20 km	100 km
Global	65000x65000x40km	1000km

- Domain** of the atmospheric model is the area that is simulated. The computation domain consists of an array of **computational cells**, each having uniform chemical composition. The size of cells determines the spatial resolution of the model.
- Atmospheric chemical transport models are also characterized by their dimensionality:
 - 0D or zero-dimensional (box) model
 - 1D or one-dimensional (column) model
 - 2D or two-dimensional model
 - 3D or three-dimensional model.

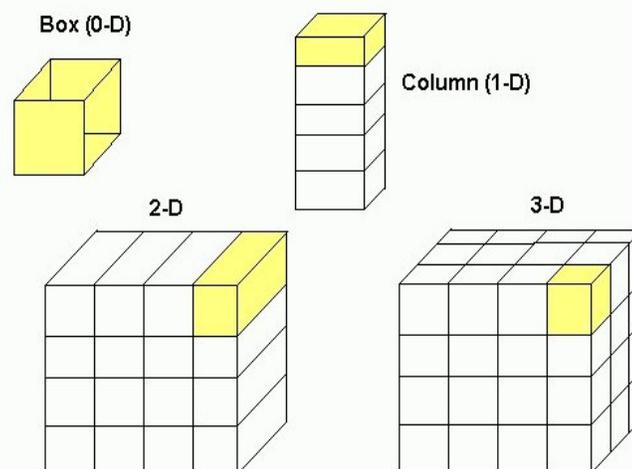


Figure 2 Schematic depiction of (a) a box model (zero-dimensional), (b) a column model (one-dimensional), (c) a two-dimensional model, and (d) a three-dimension model (from Seinfeld and Pandis, 1998).

- Model time scale depends on a specific application varying from hours (e.g., air quality model) to hundreds of years (e.g., climate models). The equations are integrated with a time step as long as possible to increase computational speed but should exceed value creating instability.

Two principal approaches to simulate changes in the chemical composition of a given air parcel:

1. Lagrangian approach:

air parcel moves with the local wind so that there is no mass exchange that is allowed to enter the air parcel and its surroundings (except of species emissions). The air parcel moves continuously, so the model simulates species concentrations at different locations at different time,

2. Eulerian approach

model simulates the species concentrations in an array of fixed computational cells.

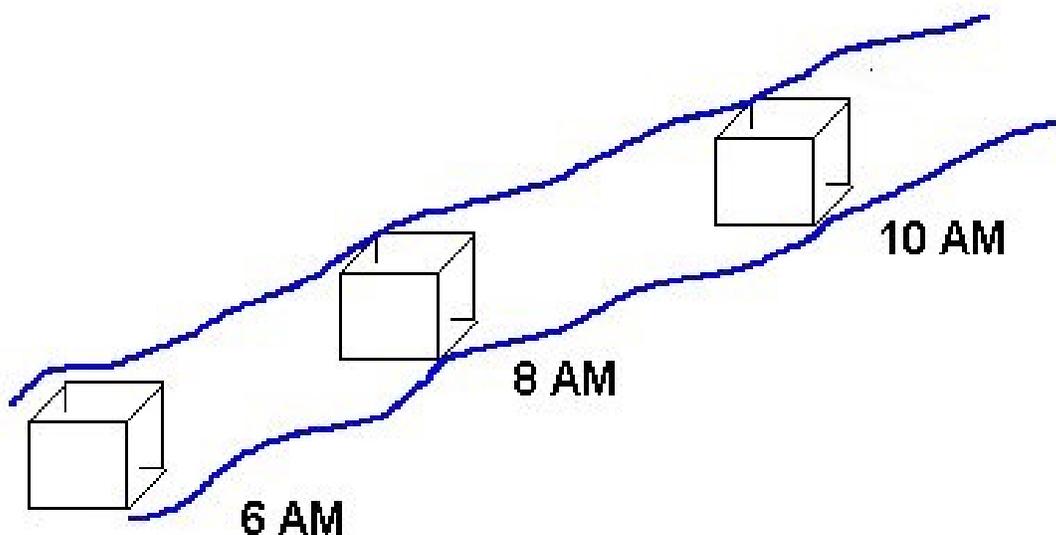


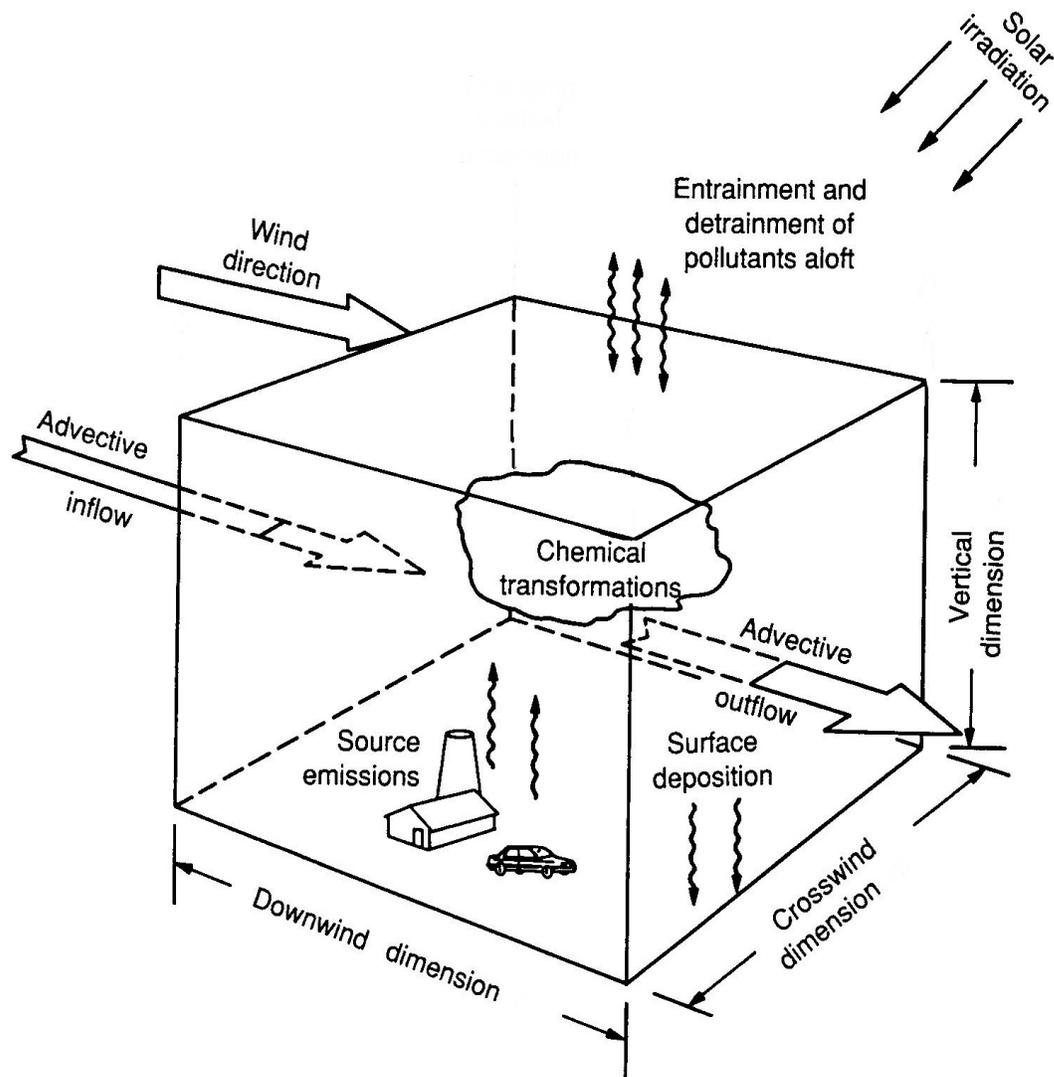
Figure 3 Movement of air parcel in a Lagrangian model.

2. Box models (or 'zero dimensional', or 0-D models)

The 0D models are the simplest models, where the atmospheric domain is represented by only one box.

- In a box model concentrations are the same everywhere and therefore are functions of time only, $n_i(t)$.

Figure 29.3 A schematic diagram of 0-D model of atmospheric chemistry (Eulerian approach).



Eulerian box model:

Aerosols enter a box in two ways:

1. source emissions;
2. transport: **advection** (the transport of a species by the mean horizontal motion of air parcel) and **entrainment** (the vertical movement of air parcels as a consequence of turbulent mixing)

Aerosols are removed from a box in three ways:

1. transport: **advection** out of the box and **detrainment** due to upwards motion;
 2. chemical transformations;
 3. removal processes: dry deposition on surface or wet deposition by precipitating clouds.
- In the Lagrangian box model: advection terms are eliminated, but source terms vary as the parcel moves over different source regions.

Some features of the box models:

- The dimensions and placement of the box is dictated by the particular problem of interest.

For instance, to study the influence of urban emissions on the chemical composition of air, the box may be design to cover the urban area.

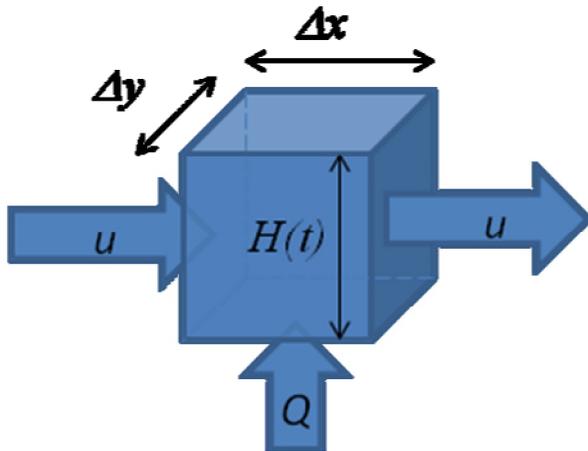
- Box models can be time dependent. In that case, any variations in time of the processes considered need to be accurately supplied to the model.

For instance, if a box model is used to compute air quality for 24-hr day in an urban area, the model must include daily variations in traffic and other sources; diurnal wind speed patterns; variations in the height of the mixed layer; variation of solar radiation during the day; etc.

Limitations of a box model:

- i. assumes rapid vertical and horizontal mixing;
- ii. assumes uniformity of surface sources;

Simplified mathematical formulation of a box model:



A separate equation is written for each chemical species of interest. For species i , present at concentration c_i , in a well-mixed box of dimension $\Delta x, \Delta y$ and height H , the time dependence of the concentration of species i is given by

$$\frac{d}{dt}(c_i \Delta x \Delta y H) = Q_i + R_i \Delta x \Delta y H - S_i + u H \Delta y (c_i^0 - c_i)$$

Where Q_i is the mass emission rate of i [$\text{kg} \cdot \text{h}^{-1}$], S_i the removal rate of i [$\text{kg} \cdot \text{h}^{-1}$], R_i its chemical production rate [$\text{kg} \cdot \text{m}^{-3} \cdot \text{h}^{-1}$], c_i^0 its background concentration, and u the wind speed.

Dividing by $\Delta x, \Delta y, H$ (assuming H constant) the box model equation becomes:

$$\frac{dc_i}{dt} = \frac{q_i}{H} + R_i - s_i + \frac{u}{\Delta x} (c_i^0 - c_i)$$

where q_i and s_i are the emission and removal rates of i per unit area [$\text{kg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$].

Dry deposition. The removal rate due to dry deposition can be described using the dry deposition velocity v_d as $s_i = v_d c_i$. The deposition process can be interpreted in terms of an electrical resistance analogy, with three resistances in series (aerodynamic resistance r_a , quasi laminar layer resistance r_b , and canopy resistance r_c) with one in parallel (gravitational settling v_s).

Assuming that particles adhere to the surface upon contact ($r_c=0$), then the vertical flux is

$$F = \frac{C_3 - C_2}{r_a} + v_s C_3 = \frac{C_2 - C_1}{r_b} + v_s C_2 = \frac{C_3 - C_1}{r_t}$$

The deposition velocity is the inverse of the total resistance r_t and is given by:

$$v_d = \frac{1}{r_a + r_b + r_a r_b v_s} + v_s$$

The box model equation becomes:

$$\frac{dc_i}{dt} = \frac{q_i}{H} + R_i - \frac{v_d}{H} c_i + \frac{u}{\Delta x} (c_i^0 - c_i)$$

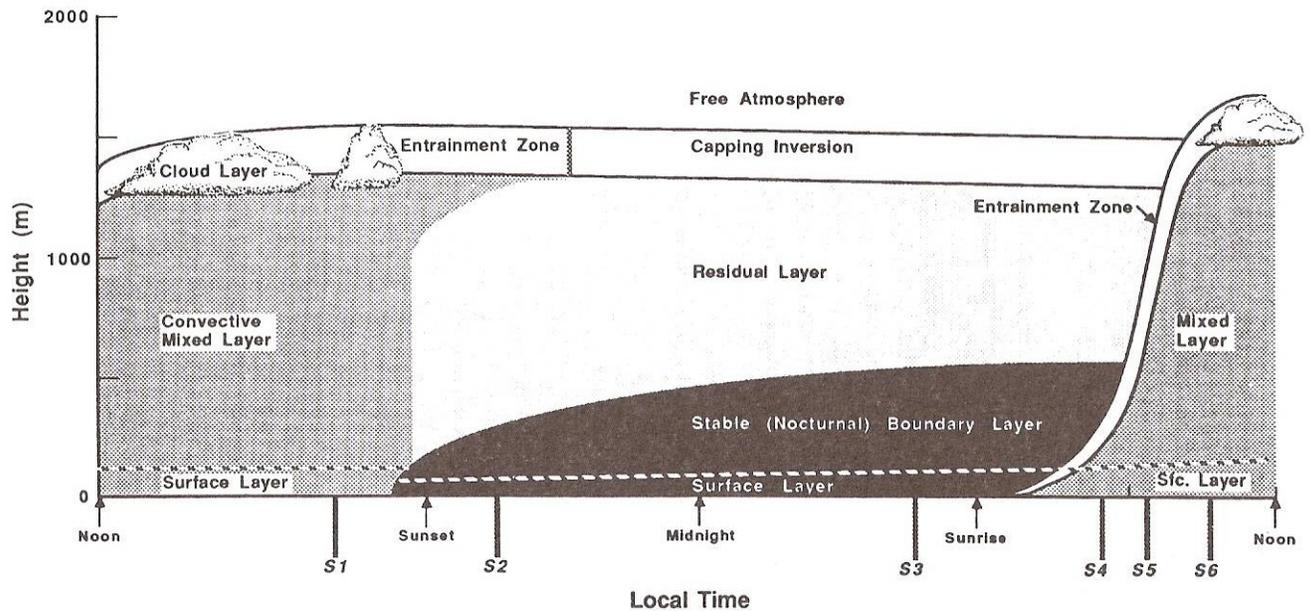
The terms in the right-hand side correspond to the changes in the concentration of I as a result of emission, chemical reaction, dry deposition, and advection.

The **residence time** of the air box advected at wind speed u over Δx is

$$\tau_R = \frac{\Delta x}{u}$$

Entrainment: The mixing height of the boundary layer varies diurnally, with low value at night and high value during the day. When the mixing height decreases there is no mixing with the environment and the concentration does not change, while the mixing height increases, there is entrainment and subsequent dilution. The increase of concentration $c_i + \Delta c_i$ by increase of mixing height $H + \Delta H$ in an environment of concentration c_i^a is given by

$$\frac{dc_i}{dt} = \frac{c_i^a - c_i}{H} \frac{dH}{dt}$$



The entraining Eulerian box model equation is:

$$\frac{dc_i}{dt} = \frac{q_i}{H(t)} + R_i - \frac{v_d}{H(t)} c_i + \frac{c_i^0 - c_i}{\tau_R} \quad \text{for } \frac{dH}{dt} \leq 0$$

$$\frac{dc_i}{dt} = \frac{q_i}{H(t)} + R_i - \frac{v_d}{H(t)} c_i + \frac{c_i^0 - c_i}{\tau_R} + \frac{c_i^a - c_i}{H(t)} \frac{dH}{dt} \quad \text{for } \frac{dH}{dt} \geq 0$$

The entraining Lagrangian box model equation is

$$\frac{dc_i}{dt} = \frac{q_i}{H(t)} + R_i - \frac{v_d}{H(t)} c_i \quad \text{for } \frac{dH}{dt} \leq 0$$

$$\frac{dc_i}{dt} = \frac{q_i}{H(t)} + R_i - \frac{v_d}{H(t)} c_i + \frac{c_i^a - c_i}{H(t)} \frac{dH}{dt} \quad \text{for } \frac{dH}{dt} \geq 0$$

- The chemical loss or production rate R_i is dependent on the number of chemical reactions included into the model, and it is given by

$$dc_i/dt = R_i = P_i - L_i c_i$$

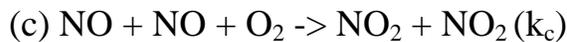
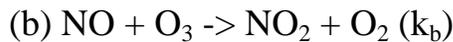
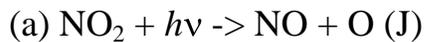
P_i is the production term of species i ; and L_i is the loss rate.

The solution of the entraining box model equation with chemical reaction is given by:

$$c_i(t) = \frac{A}{B} + (c_i(0) - \frac{A}{B})e^{-Bt} \quad \text{with } A = P + \frac{q_i}{H} + \frac{c_i^0}{\tau_R} \quad \text{and } B = L + \frac{v_d}{H} + \frac{1}{\tau_R}$$

Example of Chemical Problem:

Find the rate of change of NO₂ concentration if nitrogen dioxide is involved in the following reactions with the rate coefficients J, k_b, and k_c:



Solution:

The rates of these reactions are

$$\text{Rate}_a = J [\text{NO}_2]$$

$$\text{Rate}_b = k_b [\text{NO}] [\text{O}_3]$$

$$\text{Rate}_c = k_c [\text{NO}] [\text{NO}][\text{O}_2]$$

The rate of change of NO₂ concentration can be written as production minus loss terms. Thus,

$d [\text{NO}_2] / dt = P_{\text{NO}_2} - R_{\text{NO}_2}$ where P_{NO_2} and R_{NO_2} are total chemical production and loss terms, respectively:

$$P_{\text{NO}_2} = \text{Rate}_b + 2 \text{Rate}_c = k_b [\text{NO}] [\text{O}_3] + 2 k_c [\text{NO}] [\text{NO}][\text{O}_2]$$

$$R_{\text{NO}_2} = \text{Rate}_a = J [\text{NO}_2]$$

Therefore

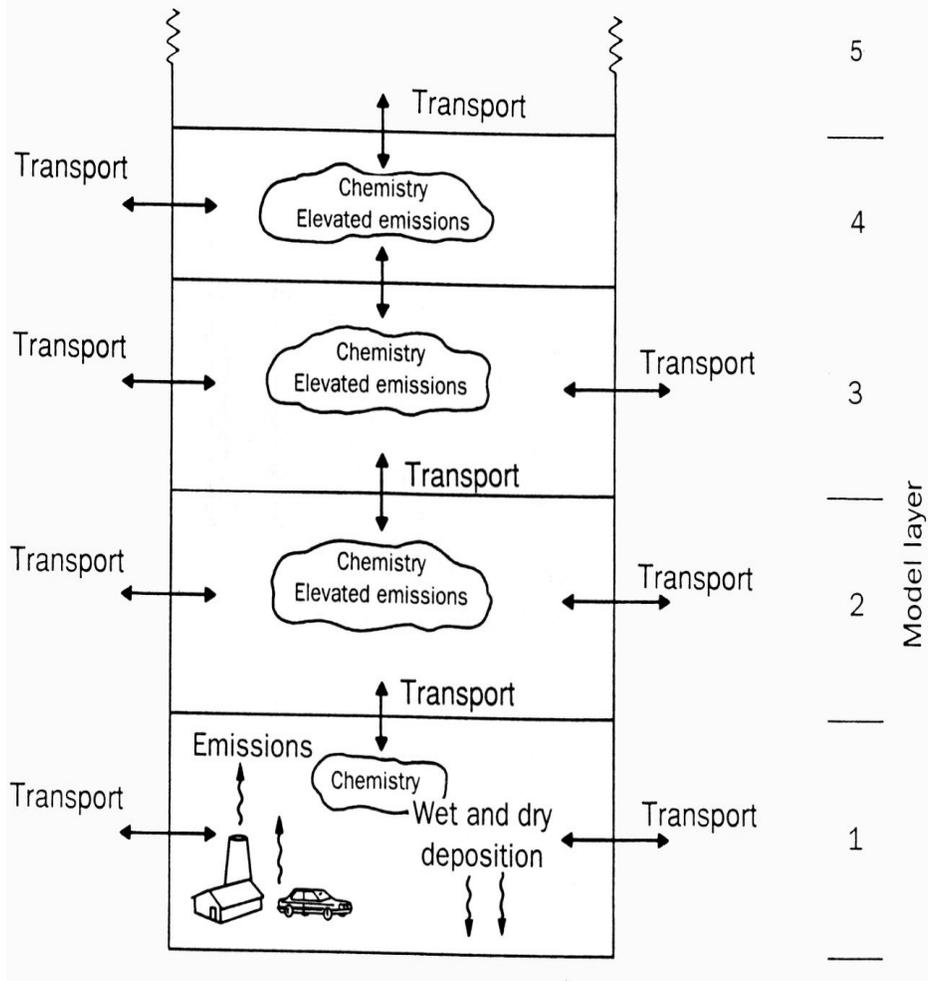
$$d [\text{NO}_2] / dt = P_{\text{NO}_2} - R_{\text{NO}_2} = k_b [\text{NO}] [\text{O}_3] + 2 k_c [\text{NO}] [\text{NO}][\text{O}_2] - J [\text{NO}_2]$$

2. One-dimensional (1-D) models

One-dimensional models assume that species concentrations are functions of height and time, $c_i(z, t)$. In these models, which ignore all variations with longitude and latitude, the exchanges of mass along the vertical are represented by an eddy diffusion formulation:

$$\frac{\partial c_i}{\partial t} + w \frac{\partial c_i}{\partial z} = \frac{1}{\rho_a} \frac{\partial}{\partial z} (K_{zz} \rho_a \frac{\partial c_i}{\partial z}) + P - Lc_i$$

where w is the vertical wind speed from large and small scale processes, K_{zz} the eddy diffusion coefficient is determined empirically.



SUBGRID PARAMETERIZATION:

A. TURBULENCE

The number of unknowns in the set of equations for turbulent flow is larger than the number of equations. For any finite set of equations the description of turbulence is not closed. The closure of turbulent

equations requires parameterizing the unknowns which cannot be solved explicitly by a prognostic equation.

Prognostic Eq. for	Order of Closure	Equation	Number of Eqs.	Number of unknowns
$\overline{U_i}$	1	$\frac{\partial \overline{U_i}}{\partial t} = \dots - \frac{\partial \overline{u'_i u'_j}}{\partial x_j}$	3	6
$\overline{u'_i u'_j}$	2	$\frac{\partial \overline{u'_i u'_j}}{\partial t} = \dots - \frac{\partial \overline{u'_i u'_j u'_k}}{\partial x_k}$	6	10
$\overline{u'_i u'_j u'_k}$	3	$\frac{\partial \overline{u'_i u'_j u'_k}}{\partial t} = \dots - \frac{\partial \overline{u'_i u'_j u'_k u'_m}}{\partial x_m}$	10	15

An example of first order closure of turbulent transport of a species i of concentration c_i consists to parameterize $\overline{w'c'_i} = -K_{zz} \frac{\partial c_i}{\partial z}$ where the parameter K_{zz} is a scalar with units of $[m^2.s^{-1}]$. Troen and Mahrt (*Bound. Layer meteor.*, 37, 129-148, 1986) have proposed the following formula for the eddy diffusion coefficient:

$$K_{zz} = k u_* z \left(1 - \frac{z}{h_{BL}}\right)^2 \quad \text{for } Ri_c < 0.25$$

$$K_{zz} = k Ri_c u_* L_{MO} \left(1 - \frac{z}{h_{BL}}\right)^2 \quad \text{for } Ri_c > 0.25$$

where $Ri_c = 0.25$ is the critical value of the Richardson Number

$$Ri_c = \frac{g \frac{\partial \theta_v}{\partial z}}{\theta_v h_{BL} \left(\frac{\partial \sqrt{u^2 + v^2}}{\partial z} \right)^2}$$

where $k=0.35$ is the von Karman constant, $g=9.807 [m^2.s^{-1}]$,

$\theta_v = T_v \left(\frac{1000}{p} \right)^{R/c_p}$ the virtual potential temperature [K],

$T_v = T(1 + 0.61q)$ the virtual temperature [K], q the specific humidity $[kg.kg^{-1}]$, $R=287 [J.kg^{-1}.K^{-1}]$ gaz constant, $c_p=1004 [J.kg^{-1}.K^{-1}]$ the specific heat capacity of air, (u,v) the horizontal wind components, h_{BL}

is the boundary layer height [m] given by $h_{BL} = \frac{Ri_c \theta_{Vs} \left| \overline{U}(h_{BL}) \right|^2}{g(\theta_v(h_{BL}) - \theta_{Vs}^*)}$, θ_{Vs}^* is

the surface virtual potential temperature in stable condition but is

$\theta_{Vs}^* = \theta_{Vs} + 6.5 \frac{\overline{(w''\theta'')}}_s}{u_*}$ for unstable condition, $u_* = \sqrt{\frac{\tau_s}{\rho}}$ is the friction velocity [m.s⁻¹], L_{MO} is the Monin-Obukhov length [m] given by

$$L_{MO} = -\frac{u_*^3}{k \frac{g}{\theta_{Vs}} \overline{(w''\theta'')}}_s$$

where $\overline{(w''\theta'')}}_s = C_h(\theta_s - \theta_0)$ is the surface heat flux [W.m⁻²], and C_h is the heat transfer coefficient [W.m⁻².K⁻¹] between the surface (subscript s) and the reference level (subscript 0). L_{MO} values show strong diurnal variation with negative values during the day, and positive at night.

B. CONVECTIVE TRANSPORT

Transport in deep convective clouds is very localized. In a region typically covering the area of a grid cell in a global model 99.9% is not affected by convective updraft.

Parameterization must be introduced to estimate the effects of these localized but intense processes on the distribution of tracers.

3. **Two-dimensional (2-D) models** assume that species concentrations are uniform along one dimension and depend on the other two and time, for example $c_i(x, z, t)$.
 - 2-D models are often used in description of global atmospheric chemistry, assuming that concentrations are functions of latitude and altitude but do not depend on longitude.
 - 2-D models have mainly been used for stratospheric studies, because in troposphere there are essential longitudinal variations.

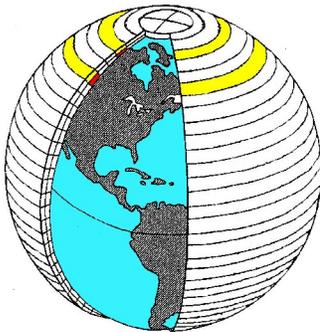


Figure 4 Computational cells of 2D model

3. **Three-dimensional (3-D)** models simulate the full concentration field $c_i(x, y, z, t)$.

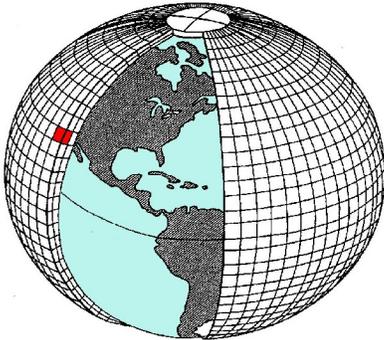


Figure 5 Computational cells of 3D model.

- 3-D models require simultaneous calculations of the meteorological field and chemistry. The chemistry is often very simplified, because of computational complexities of the meteorological field.

Decoupled (or off-line) 3-D chemical model:

first meteorological model is run to for chosen time period and for a geographical location (or entire globe), and the calculated wind field, temperature, water vapor, cloud, etc. are stored to be used as input variable in 3-D chemical model.

Advantage of decoupled 3-D model: treats photochemical processes with more details.

- 3-D chemical transport model seeks a solution of the **continuity (or mass conservation) equation** for each species (gas or aerosols) included in the model.

General form of the continuity equation:

$$dc_i/dt = (dc_i/dt)_{\text{tran}} + (dc_i/dt)_{\text{cloud}} + (dc_i/dt)_{\text{rem}} + (dc_i/dt)_{\text{aeros}} + (dc_i/dt)_{\text{chem}} + S_i$$

where c_i is the concentration of a species i ;

$(dc_i/dt)_{\text{tran}}$ is the rate of change of c_i due to transport (advection, diffusion, etc.);

$(dc_i/dt)_{\text{cloud}}$ is the rate of change of c_i due to cloud processes (cloud scavenging, evaporation of cloud droplets, aqueous-phase reactions, wet deposition, etc.);

$(dc_i/dt)_{\text{aeros}}$ is the rate of change of c_i due to aerosol processes (transport between gas and aerosol phases, aerosol microphysical transformations, etc.);

$(dc_i/dt)_{\text{chem}}$ is the rate of change of c_i due to gas-phase reactions;

Q_i is the emission rate.

Which models are used for which applications:

0-D and 1-D models are used when one knows very little about a problem or when data are not available to validate model performance;

2-D models are used when a horizontal dimension is important.

3-D models are used when the most complete answers are required, but computer resources and model building still severely limit their use.