Atmospheric Chemical Transport Modelling:

Basic Processes

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(Using some materials of previous School lecture by M. Sofiev, FMI, and DMI team contributions)

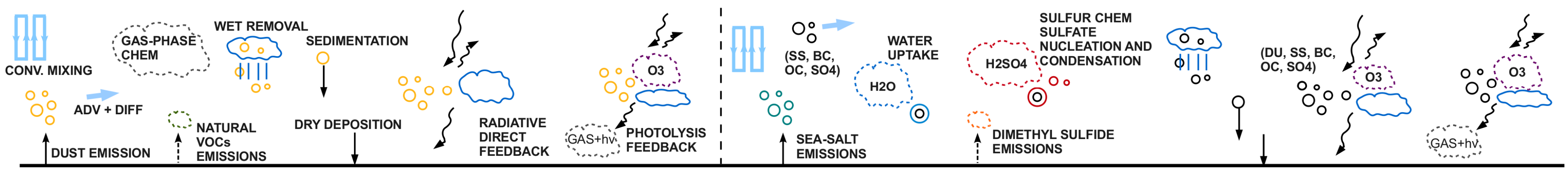
MUSCATEN summer school
"Integrated Modelling of Meteorological and Chemical Transport Processes / Impact of Chemical Weather on Numerical Weather Prediction and Climate Modelling"

Odessa, Ukraine, 3-9 July 2011

Lecture's Objective and Goal:

- ***General Introduction to the Physical and Chemical Atmospheric Processes & Physical Atmospheric Processes, characteristics of atmospheric composition and air quality, model evaluation.***

- The basic Atmospheric Chemical Transport Modelling (ACTM) processes are shortly introduced.
- This includes: advection, diffusion, deposition, emission, chemistry, aerosols, and clouds. *These processes will be handled in more details in the following lectures.*
- Definitions, diffusion, deposition and land use. How are they solved in CTM.
- Means of characterization of atmospheric composition, appropriate measures and consequences for the CTM evaluation.



Jorba, EGU-2011

Table 3.4. Some Gases and Aerosol Particle Components Important for Specified Air Pollution Topics

Indoor Air Pollution	Outdoor Urban Air Pollution	Acid Deposition	Stratospheric Ozone Reduction	Global Climate Change
Gases				
Nitrogen dioxide Carbon monoxide Formaldehyde Sulfur dioxide Organic gases Radon	Ozone Nitric oxide Nitrogen dioxide Carbon monoxide Ethene Toluene Xylene PAN	Sulfur dioxide Sulfuric acid Nitrogen dioxide Nitric acid Hydrochloric acid Carbon dioxide	Ozone Nitric oxide Nitric acid Hydrochloric acid Chlorine nitrate CFC-11 CFC-12	Water vapor Carbon dioxide Methane Nitrous oxide Ozone CFC-11 CFC-12
Aerosol Particle Components				
Black carbon Organic matter Sulfate Nitrate Ammonium Allergens Asbestos Fungal spores Pollens Tobacco smoke	Black carbon Organic matter Sulfate Nitrate Ammonium Soil dust Sea spray Tire particles Lead	Sulfate Nitrate Chloride	Chloride Sulfate Nitrate	Black carbon Organic matter Sulfate Nitrate Ammonium Soil dust Sea spray

(after Jacobson, 2002)

Urban Air Chemistry

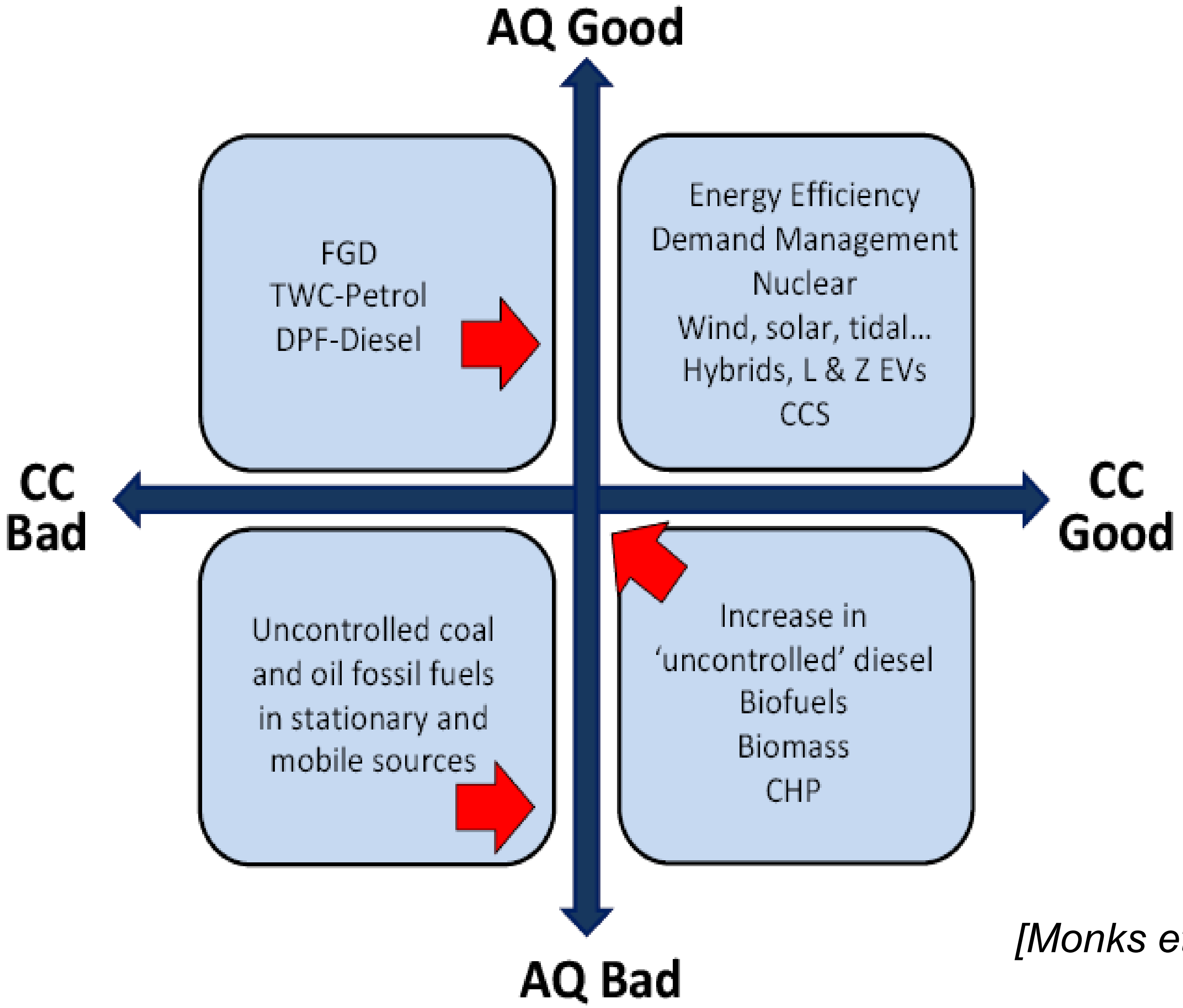
Primary Emissions	Sources	Secondary Pollutants	Effects
Sulfur Dioxide (SO_2)	Power Plants, Vehicles, Industry	Sulfuric Acid Vapor, Sulfate Particles	Acid Rain, Heart/Lung Disease, Haze, Climate Change
Nitrogen Oxides (NO_x)	Power Plants, Vehicles, Trash Burning, Industry	Nitric Acid Vapor, Nitrate Particles	Acid Rain, Heart/Lung Disease, Smog, Haze, Climate Change
Volatile Organic Compounds (VOCs)	Vehicles, Industry, Painting, Cleaning, Cooking	Organic Air Toxics, Organic Particles	Smog, Heart/Lung Disease, Haze, Climate Change
Ammonia (NH_3)	Vehicles, Human and Animal Waste	Ammonium Sulfate, Nitrate, Particles	Haze, Heart/Lung Disease, Climate Change

Effects of degraded air quality in cities

- **Adverse health impacts**
- **Visibility impairment**
- **Regional ecosystem impacts**
  - Acid and fixed nitrogen deposition
  - Photochemical oxidant damage
  - Photosynthetically active radiation
- **Regional/urban weather and climate change**
- **Global pollutant transport**

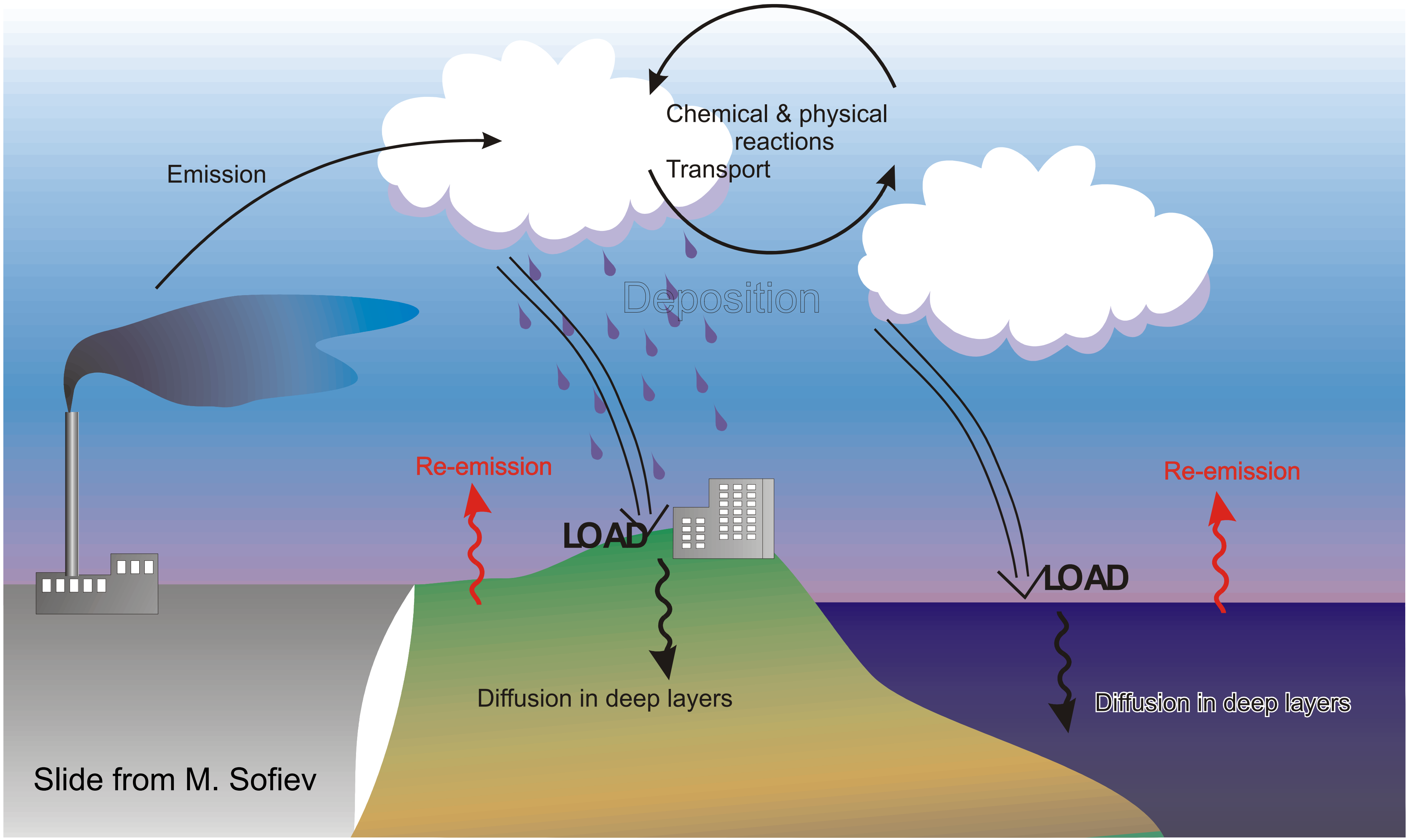
(Molina, 2004)

Synergies and trade-offs between policies to improve air quality and to reduce greenhouse gas emissions



[Monks et al., 2009]

Pollution cycle in the troposphere



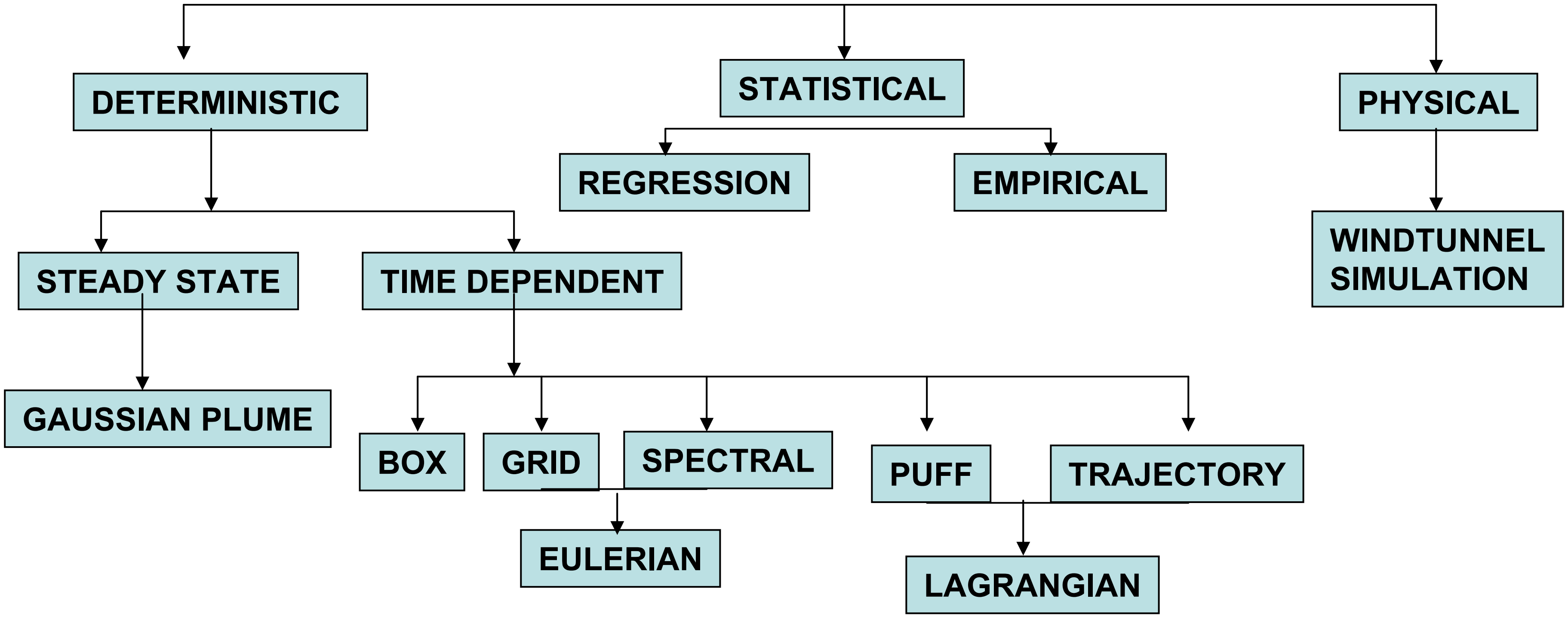
ACT models consist of the following modules:

- *Transport and diffusion*—calculating three-dimensional motion of gases and aerosols in a gridded model domain
- *Gas-phase chemistry*—calculating changes in gaseous concentrations due to chemical transformations
- *Aerosol*—calculating size distribution and chemical composition of aerosols accounting for chemical and physical transformations
- *Cloud/fog meteorology*—calculating physical characteristics of clouds and fog based on the information from the meteorological model (or from observations)
- *Cloud/fog chemistry*—calculating changes in chemical concentrations in clouds/fog water
- *Wet deposition*—calculating the rates of deposition due to precipitation (and, possibly, cloud impaction and fog settling) and the corresponding changes in chemical concentrations
- *Dry deposition*—calculating the rates of dry deposition for gases and aerosols and the corresponding changes in their concentrations

Classifications of AQ Models

- Developed for a number of pollutant types and time periods
  - **Short-term** models – for a few hours to a few days; worst case episode conditions
  - **Long-term** models – to predict seasonal or annual average concentrations; health effects due to exposure
- Classified by
  - **Non-reactive** models – pollutants such as SO_2 and CO
  - **Reactive** models – pollutants such as O_3, NO_2, etc.
- Classified by coordinate system used
  - **Grid-based**
    - Region divided into an array of cells
  - **Trajectory**
    - Follow plume as it moves downwind
- Classified by level of sophistication

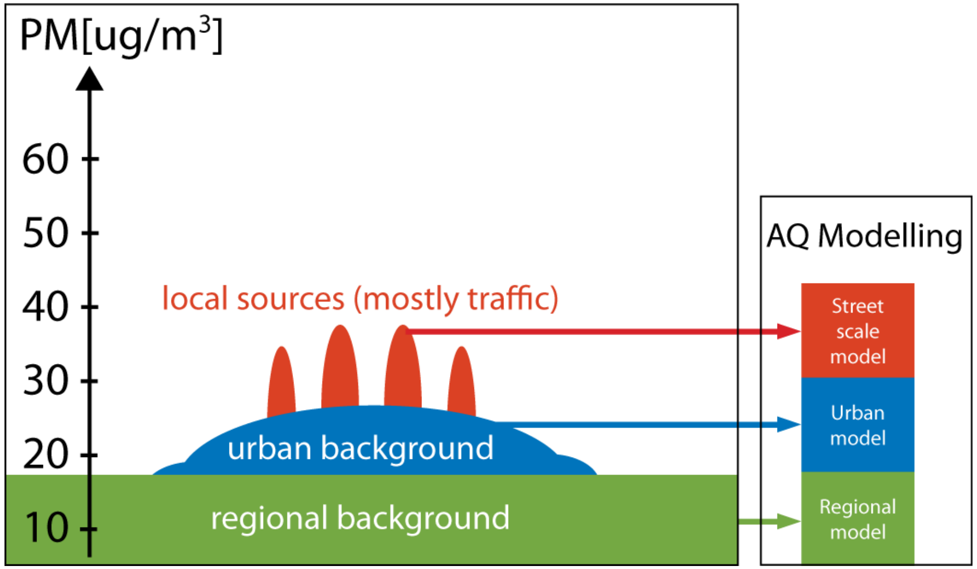
Air Quality Models



Weber, 1982

Scales of atmospheric composition

- A specific feature of the atmospheric composition problem is a very wide range of scales, both temporal and spatial combined with very sharp gradients of the species
  - *scales are largely dictated by chemical and removal lifetimes*
  - *gradients are largely dictated by sources*
- Gradients tend to reproduce themselves at every spatial scale, from street-canyon to global
  - *consequence: at every resolution the model has to be able to deal with highly irregular field*
- Non-linearities in the governing equations make averaging problematic and further complicate the scale interaction problem.

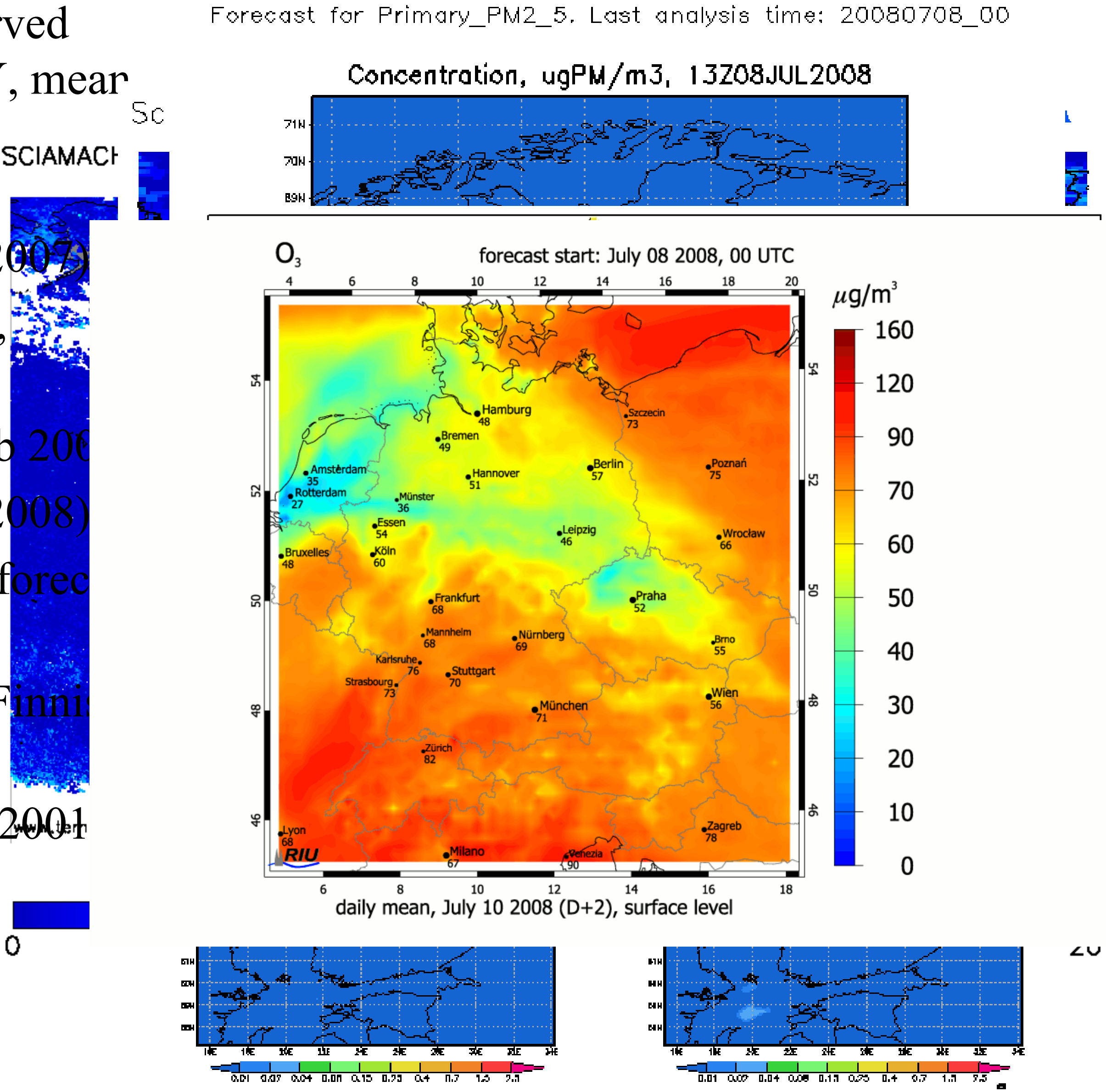


ACCENT, 2009

Up- and Down-scaling methodologies:
- Advanced coupling schemes
- Simpler schemes

Scales of atmospheric composition - 2

- Global NO2 in column observed from space (SCHIAMACHY, mear July 2007)
- NO2 column over Europe (SCIAMACHY, mean July 2007)
- PM 2.5 observed from space, Northern Italy (June 2004)
- Global CO, modelled (17 Feb 200
- NO_2 forecast, Europe (10.7.2008)
- Ozone over Central Europe, forec (8.7.2008)
- Ex.2: Primary PM 2.5 from Finni sources, forecast (8.7.2008)
- Ex.4 NO2 for Lisbon (mean 2001 2002)

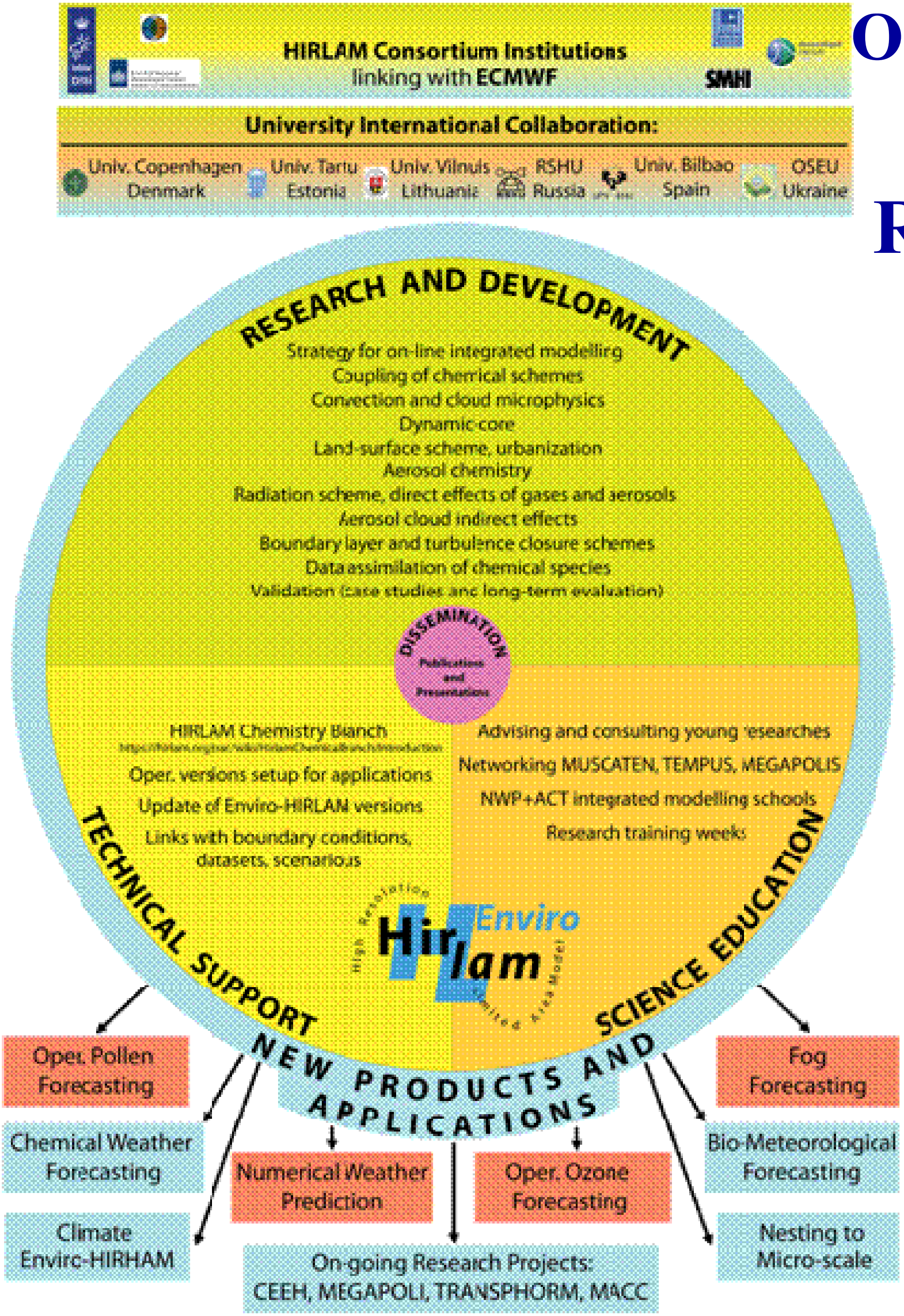


Slide from M. Sofiev

Key parameters for urban models of different scales (COST715)

Mesoscale models	Sub-meso scale models	Street canyon scale models
z_0, z_{0T}	$z_0(x)$, d(x)	
h_{UBL}	L_c, L_a, z_*	Detailed geometry
'Surface' fluxes (effective)	u_*^{IS}, H^{IS}, general: x_*^{IS}	$\bar{u}(h)$ second velocity scale for **horizontal** transport
Anthropogenic heat flux (non-surface) at some representative height	Dispersive fluxes	Heat exchange at vertical and horizontal building surfaces
Profiles of turbulent fluxes	Profiles of turbulent fluxes	Characteristic velocity variance in street canyon
Higher order moments?	Higher order moments (skewness, ...)	Higher order moments?
Synoptic forcing, average albedo	Mesoscale stability, albedo(x)	

Online coupled NWP-ACT Enviro-HIRLAM: Regional to City-Scale

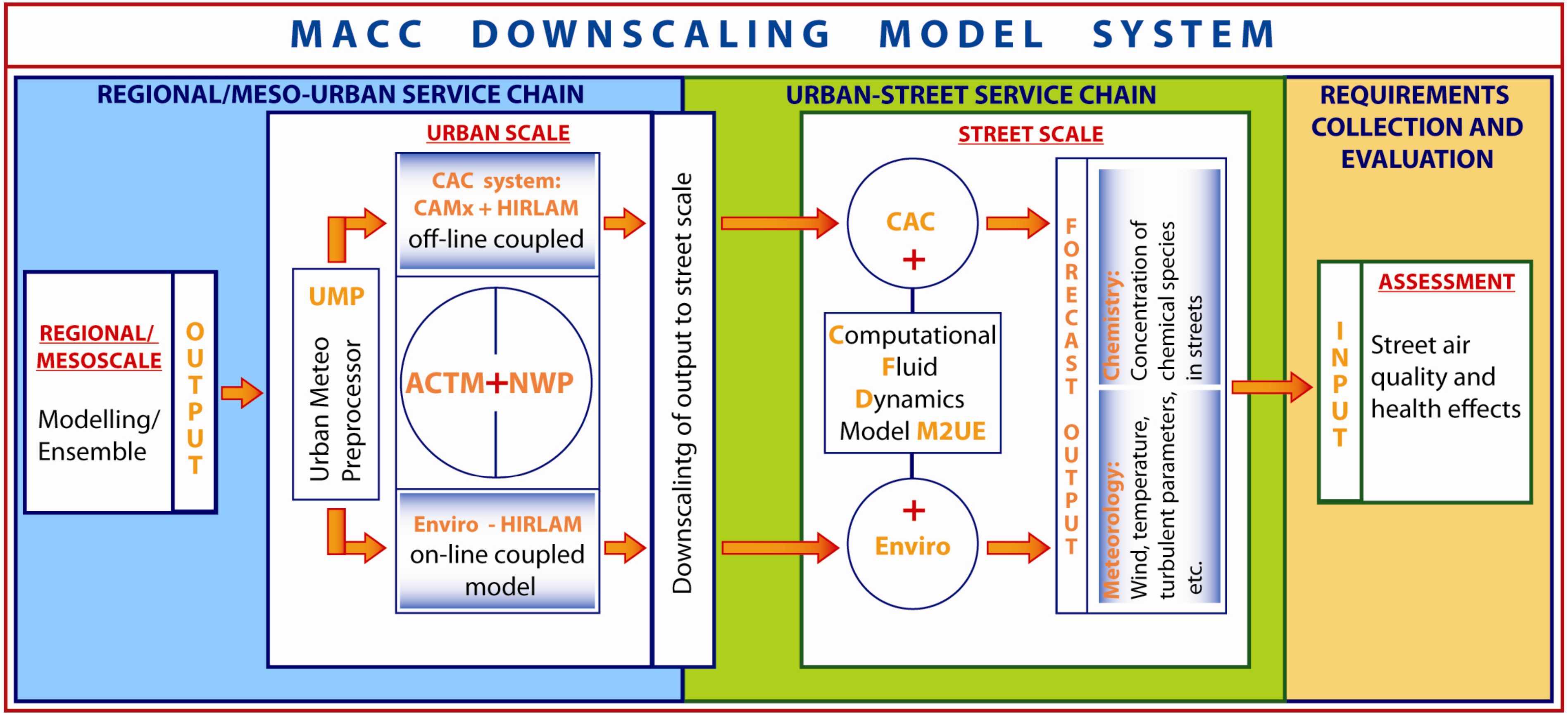


Modelling System:
On-line Enviro-HIRLAM

Global Scale Forecast:
from IFS and MACC
Regional Scale Forecast:
from DMI-HIRLAM & GEMS/MACC R-ENS

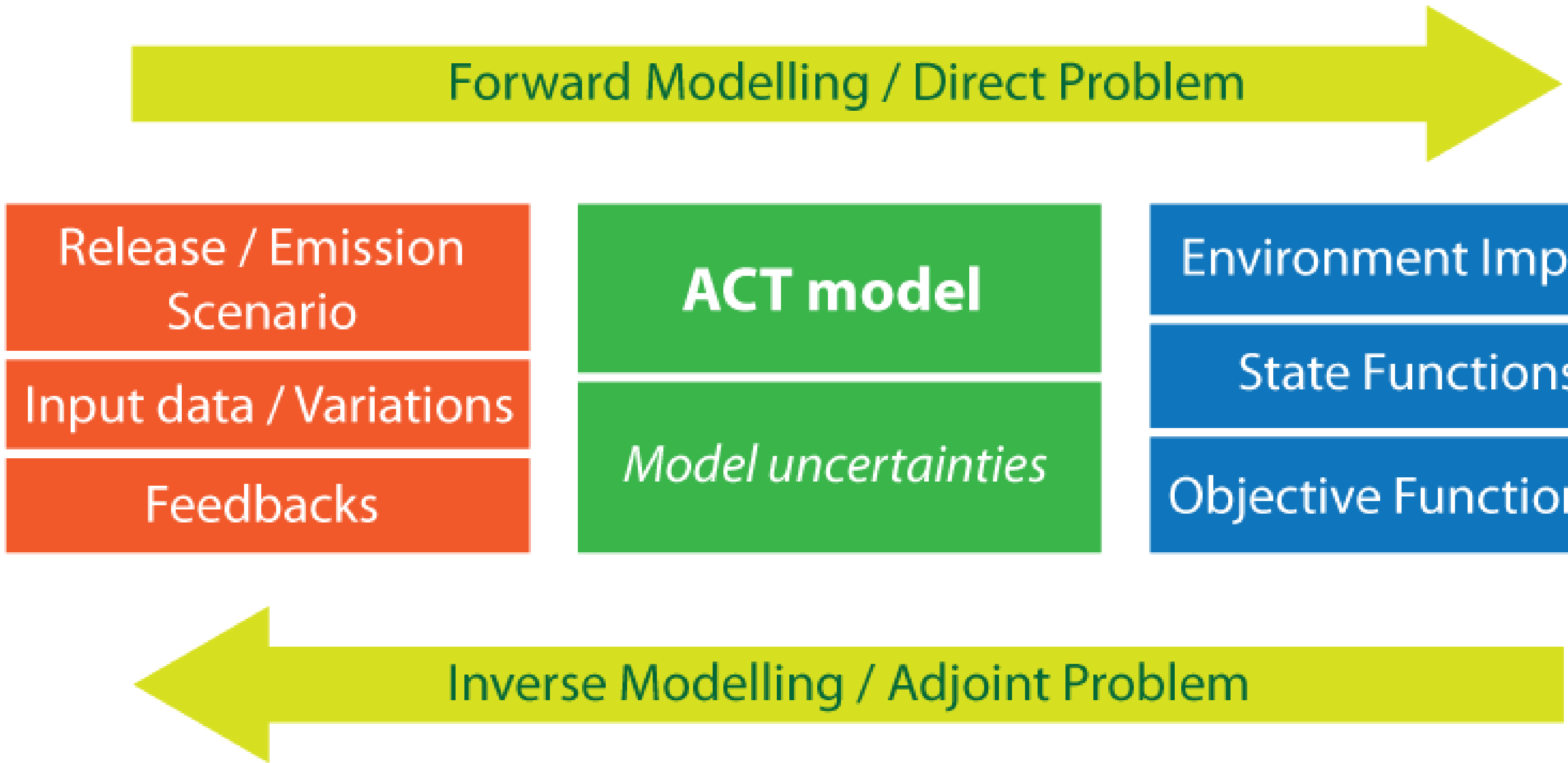
Nesting to Domains:
Regional: Europe, Denmark, …
City-scale: Copenhagen, Paris, St-Petersburg, Bilbao, Vilnus, …

Core-Downstream Modelling Chain



Downscaling of European-scale forecasts for the city and streets in Copenhagen, DK

Environmental risk assessment and mitigation strategy optimization basing on ACT model and forward/inverse modelling



Basic equations

- ***Eulerian approach***

advection — diffusion — chemistry removal — emission

$$L\overline{\varphi} \equiv \frac{\partial \overline{\varphi}}{\partial t} + \frac{\partial}{\partial x_i}(\overline{u}_i \overline{\varphi}) + \frac{\partial}{\partial x_i}(\overline{u_i^{'} \varphi^{'}}) + \sigma(\overline{\varphi}) = \bar{f}$$

$$u_i = \overline{u}_i + u_i^{'} \; : \; turbulent \;\; wind = mean + fluctuation$$

$$\varphi = \overline{\varphi} + \varphi^{'}$$

$$\frac{\partial u_i}{\partial x_i} = 0 \;\; : \;\; continuity \;\; equation$$

Basic equations - 2

K-theory: $\overline{u_i' \varphi'} \approx -\mu_{ij} \dfrac{\partial \overline{\varphi}}{\partial x_j}$

$$L\varphi \equiv \frac{\partial \varphi}{\partial t} + \frac{\partial}{\partial x_i}(u_i \varphi) - \frac{\partial}{\partial x_i} \mu_i \frac{\partial \varphi}{\partial x_i} + \sigma \varphi = f$$

Boundary & initial conditions:

$$\varphi(t = 0) = \varphi_0$$

$$\varphi(\vec{r} \in S) = \varphi_S$$

Basic equations - 3

- Assumptions and simplifications for specific AQ models
- Typical mathematical formulation of air pollution model:

Advection

Diffusion

Emission and wet deposition

Chemistry

$$\frac{\partial c_i}{\partial t} = -\left(u\frac{\partial c_i}{\partial x} + v\frac{\partial c_i}{\partial y} + w\frac{\partial c_i}{\partial z} \right)$$
$$+ K_x \frac{\partial^2 c_i}{\partial x^2} + K_y \frac{\partial^2 c_i}{\partial y^2} + \frac{\partial}{\partial z}\left(K_z \frac{\partial c_i}{\partial z} \right)$$
$$+ E_i(x, y, z, t) - \Lambda c_i$$
$$+ Q_i(c_1, c_2, \ldots, c_q)$$

J. Christensen

Basic equations - 4

Forward problem:

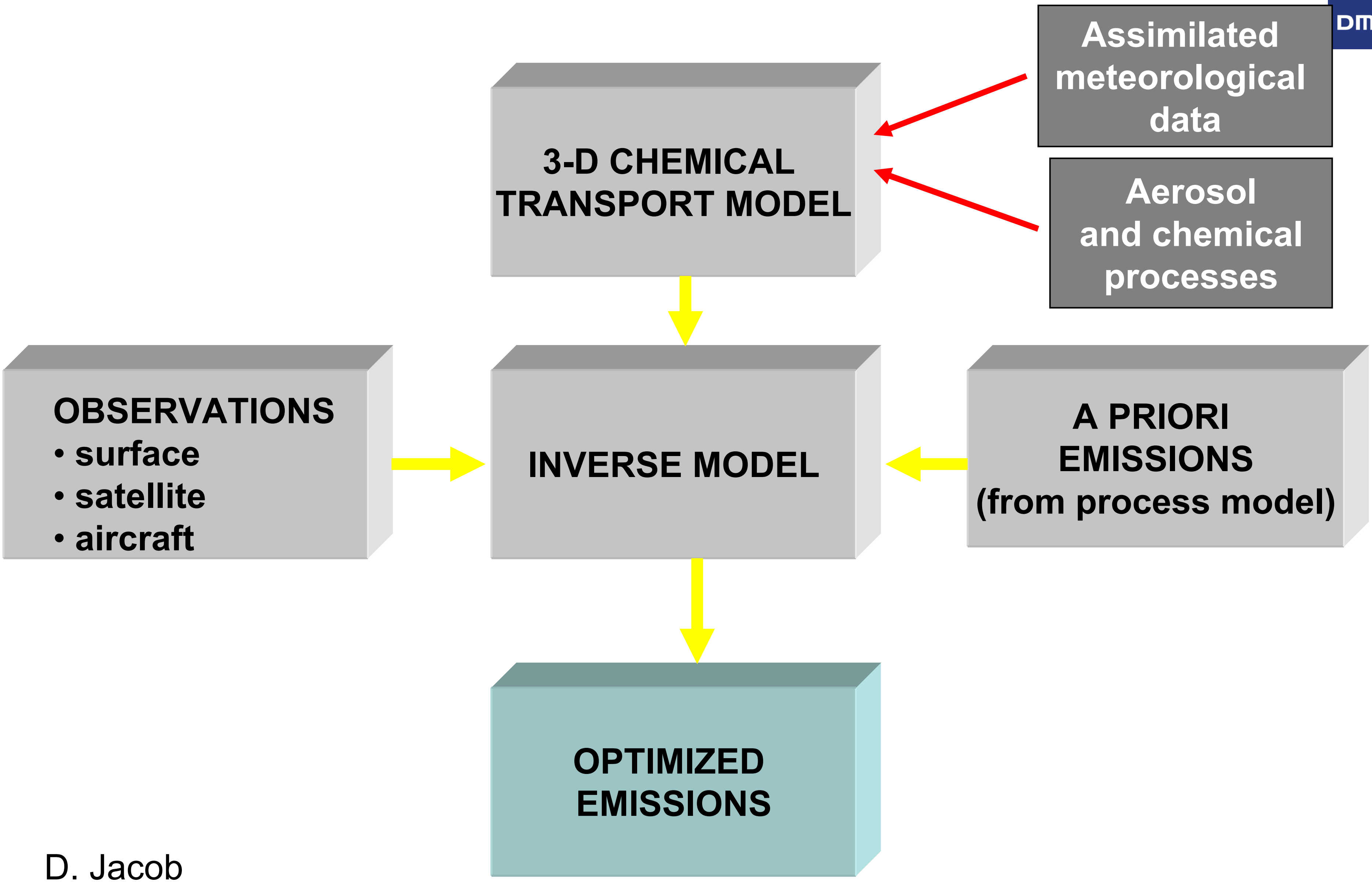
$$L = \frac{\partial}{\partial t} + \frac{\partial}{\partial x_i}(u_i) - \frac{\partial}{\partial x_i}\mu_i\frac{\partial}{\partial x_i} + \sigma; \qquad L\varphi = f; \;\; M = (p, \varphi)$$

Inverse (adjoint) problem

$$L^* = -\frac{\partial}{\partial t} - \frac{\partial}{\partial x_i}(u_i) - \frac{\partial}{\partial x_i}\mu_i\frac{\partial}{\partial x_i} + \sigma; \qquad L^*\varphi^* = p; \;\; M = (f, \varphi^*)$$

advection — diffusion — chemistry removal — receptor sensitivity — emission

Future monitoring of emissions by inverse modeling of atmospheric observations



D. Jacob

Operator splitting in Eulerian models

Reduces dimensionality of problem

Physical processes-based split (locally independent, additive processes):

$$\frac{\partial n}{\partial t} = \left[\frac{\partial n}{\partial t}\right]_{TRANSPORT} + \left[\frac{dn}{dt}\right]_{CHEMISTRY}$$

Transport $\equiv$ advection, convection: $\left[\frac{dn}{dt}\right]_{TRANSPORT} = -\nabla \bullet (n\mathbf{U})$

Chemistry $\equiv$ chemistry, emission, deposition, aerosol processes:

$$\left[\frac{dn}{dt}\right]_{CHEMISTRY} = P - L$$

Integrate each process separately over discrete time steps:

$$n(t_o + \Delta t) = \mathbf{C} \bullet \mathbf{T}\left(n(t_o)\right)$$

These operators can be split further:

- **split transport into 1-D advective and turbulent transport for *x, y, z* (usually necessary)**
- **split local into chemistry, emissions, deposition (usually not necessary)**
- **symmetrization of the algorithms within a single time step**

Structure of an ACT model

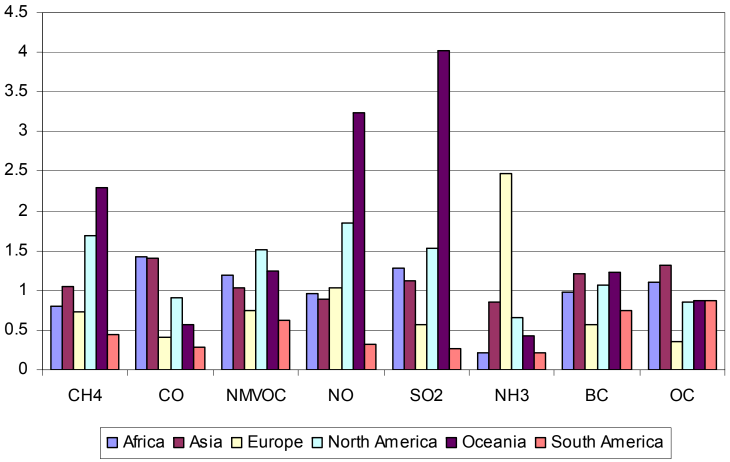
- Input data pre-processors
  - emission
  - meteorology
  - physiography (domain properties)
- Dynamic emission (simulated vs imported)
- Advection scheme
- Diffusion module
- Chemical transformation module
- Aerosol dynamics module
- Dry and wet deposition module
- Diagnostic quantities
- Output post-processing

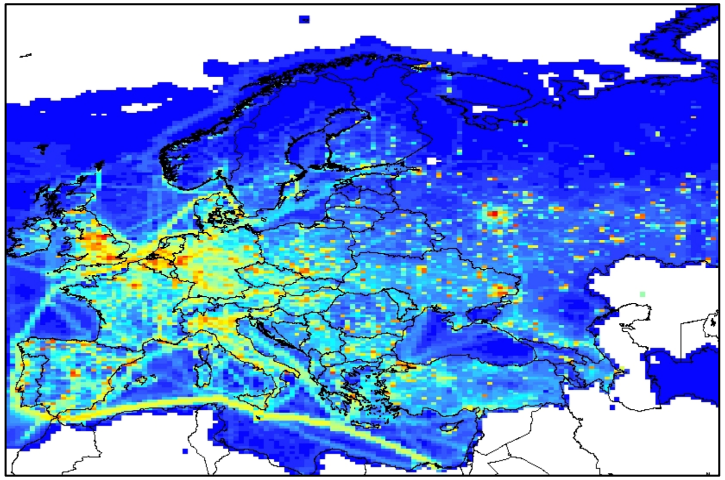
Input data pre-processors

- Emission
  - various source types (point, area, stack…)
  - time variation (diurnal, weekly, seasonal)
  - chemical content (time-dependent)
  - meteorology dependent emissions (pollen, etc)
- Meteorology (only for off-line models)
  - create extra variables (e.g. ABL parameters)
  - interpolation to the model grid
  - time interpolation

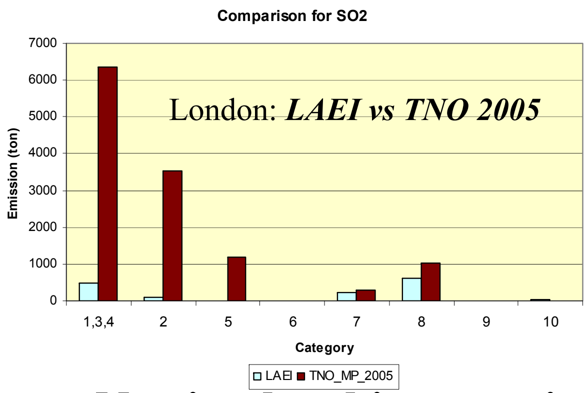
Emission Inventories

Lead by TNO Team: H. Denier van der Gone et al.

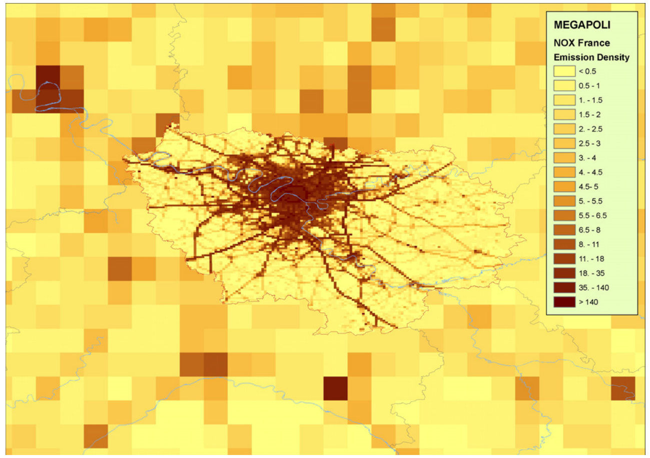




Emissions per capita in megacities (relative compared to all megacities)

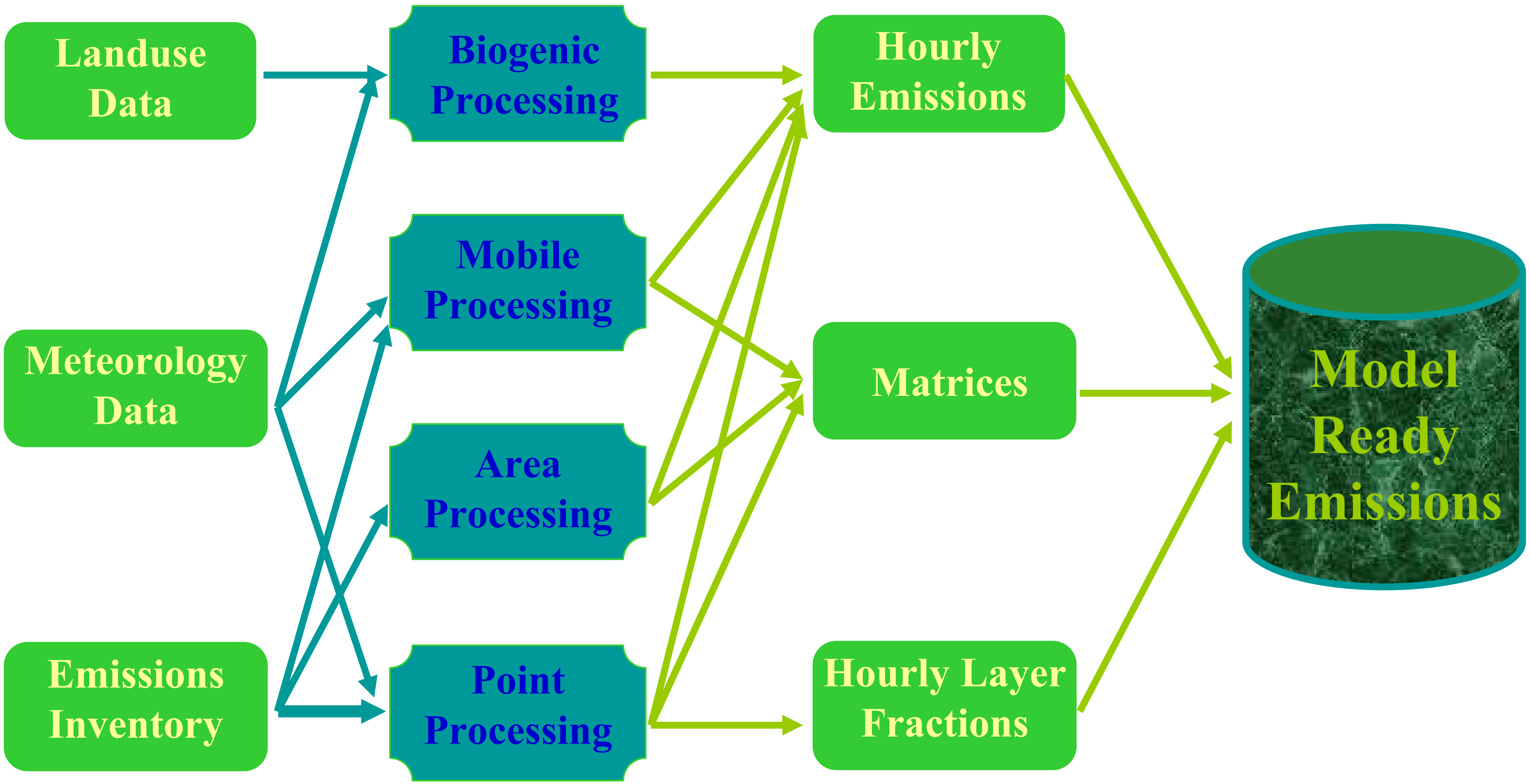


- **High uncertainties!**
- **Anthropogenic emissions**
- **Global emission inventory with all megacities mask**
- **2005 European emission inventory (6x6 km resolution)**
- **Natural emissions (e.g., fire, sea salt, pollen, volcano)**



- **Nesting local inventories for 4 megacities in focus (up to 1x1 km resolution)**
- **Comparison of city, national and European emissions**
- **Integration of nested emissions into multi-scale modelling chain**
- **European and megacity baseline scenarios for 2020, 2030 and 2050 (USTUTT)**

Emission Model (SMOKE)



(SMOKE: Sparse Matrix Operator Kernel Emissions modeling system)

Calculating the emissions per grid-box

$$M(X)_m = \sum_{k=1}^{n} EF_k(X) \times A_m \times \beta_k \times AFL_k$$

$$AFL_k = \sum_{t=1}^{9} fc_t \times \sum_{p=1}^{5} \chi_{t,p} \times m_{t,p}$$

M (X) $_m$: amount of species X emitted per month **m**

n: number of ecosystems (5)

EF $_k$ **(X)**: emission factor for species X per ecosystem

A $_m$: area burnt per month

β $_k$: combustion efficiency for ecosystem k

AFL $_k$: available fuel load per

fc $_t$: fractional cover of PFT t per gridbox

t: number of PFT's (9)

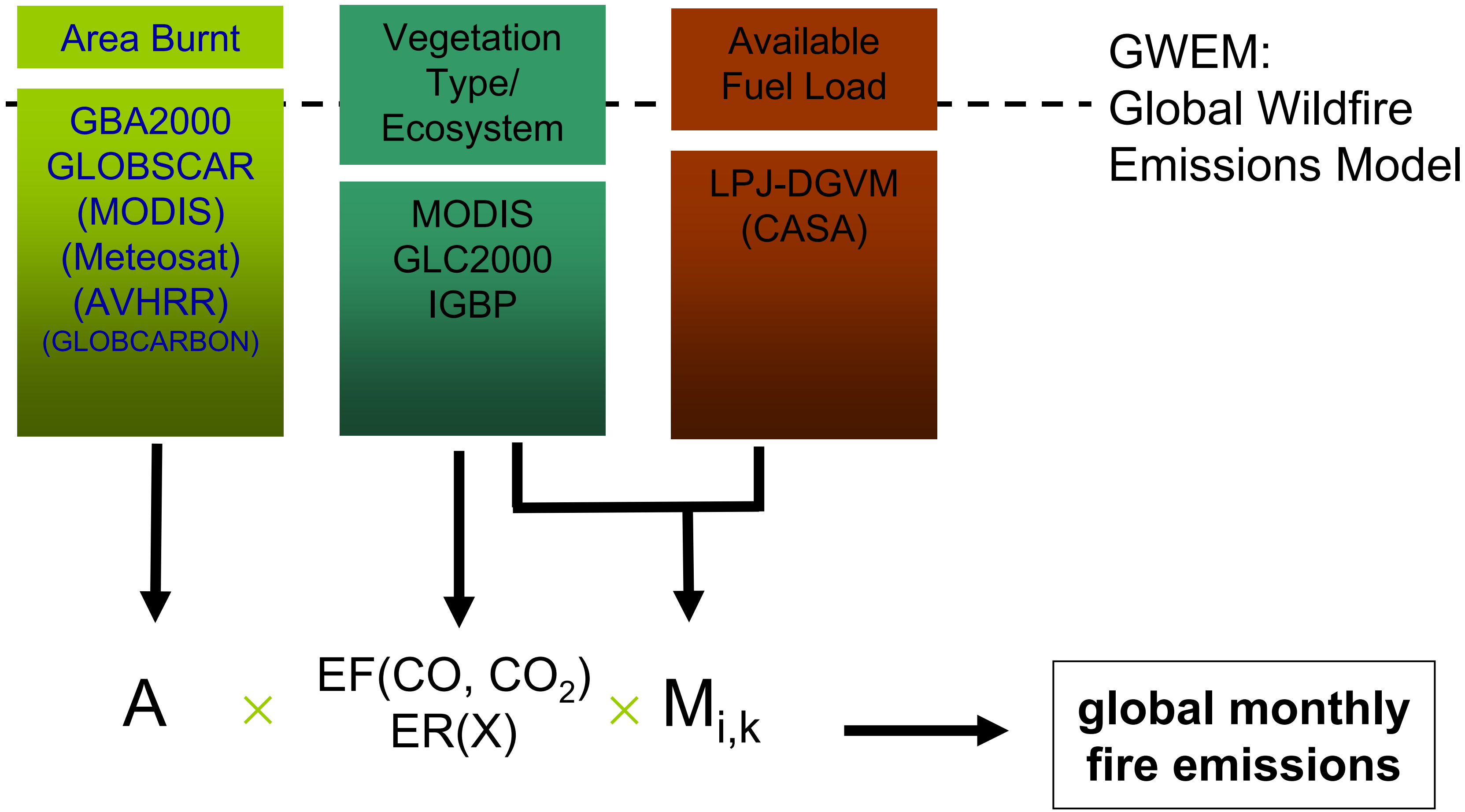
p: number of carbon pools (5)

$\chi_{t,p}$: susceptibility factor

m $_{t,p}$: dry matter per PFT and carbon pool

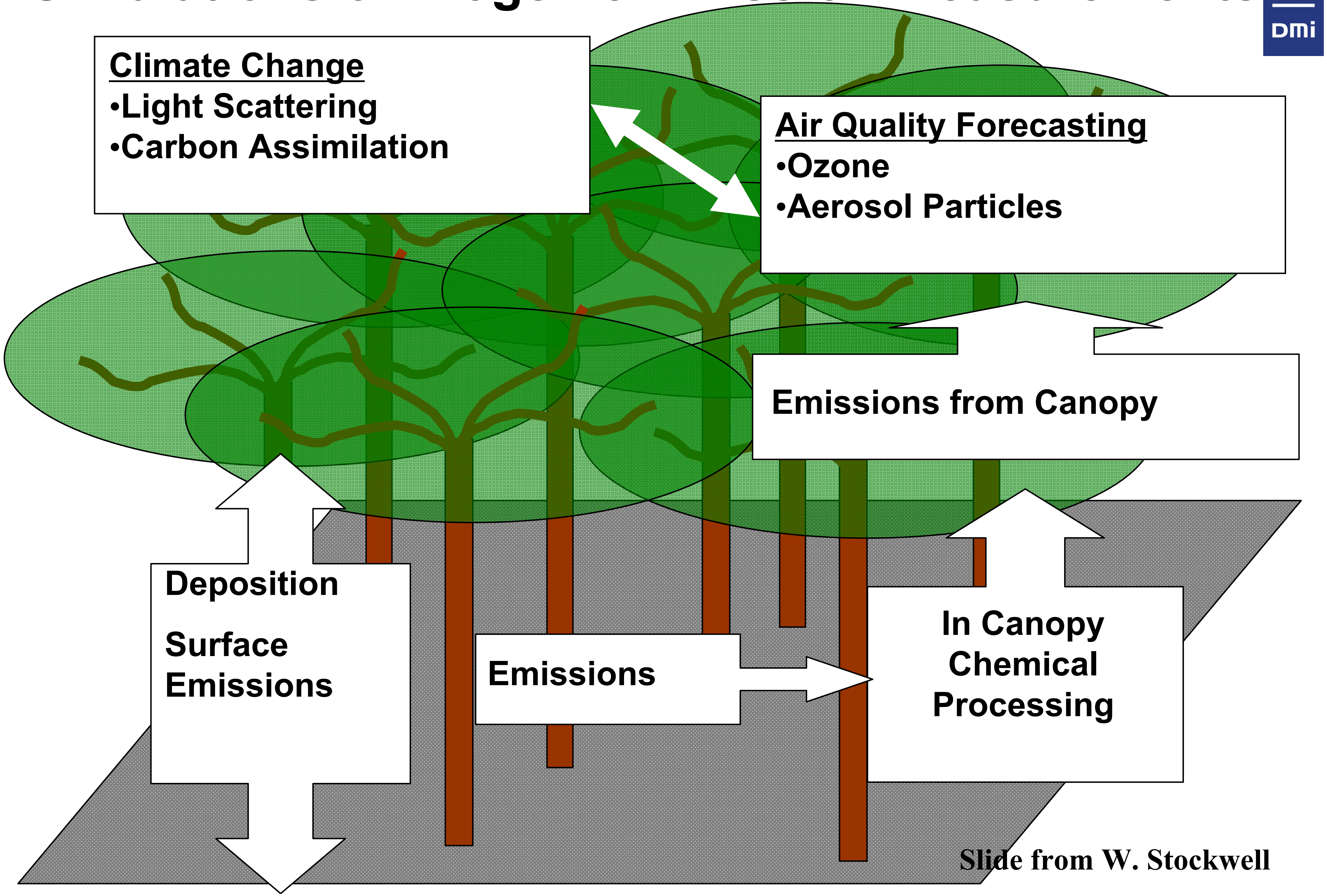
Slide from A. Zakey, R. Nuterman

Estimating Fire Emissions



Slide from A. Zakey

Biosphere - Atmospheric Interactions
Simulations of Biogenic Emission Measurements

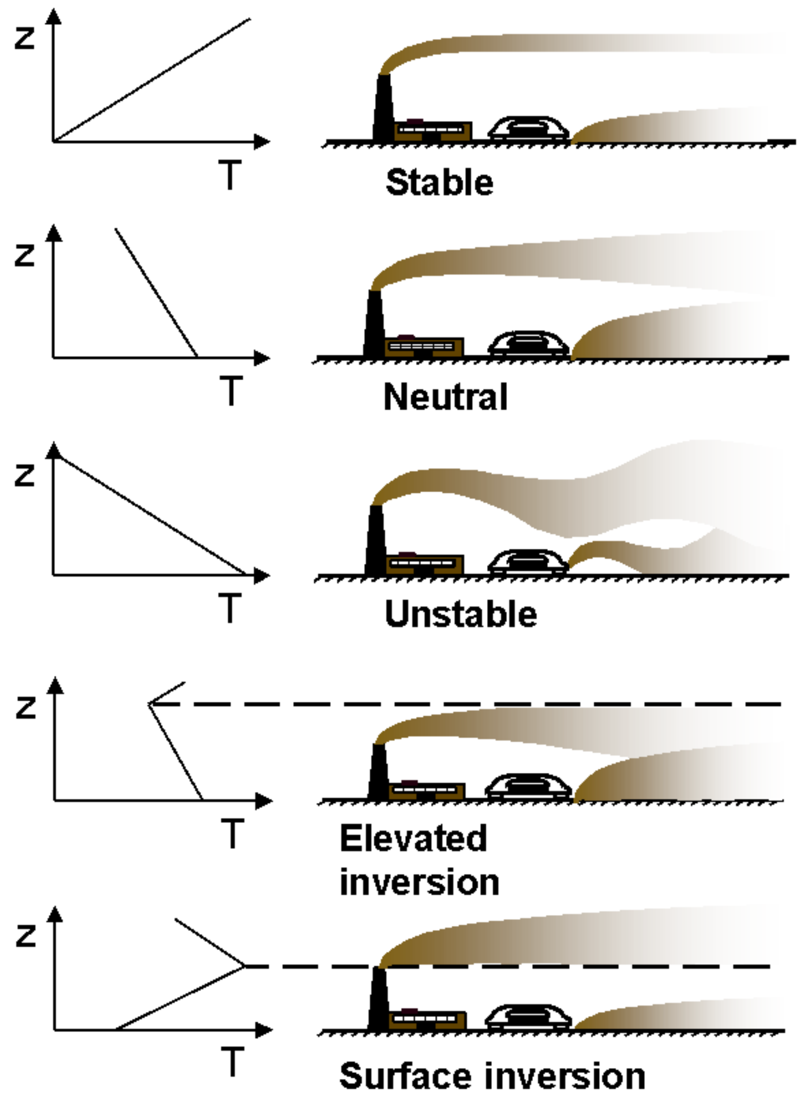


Slide from W. Stockwell

Diffusion

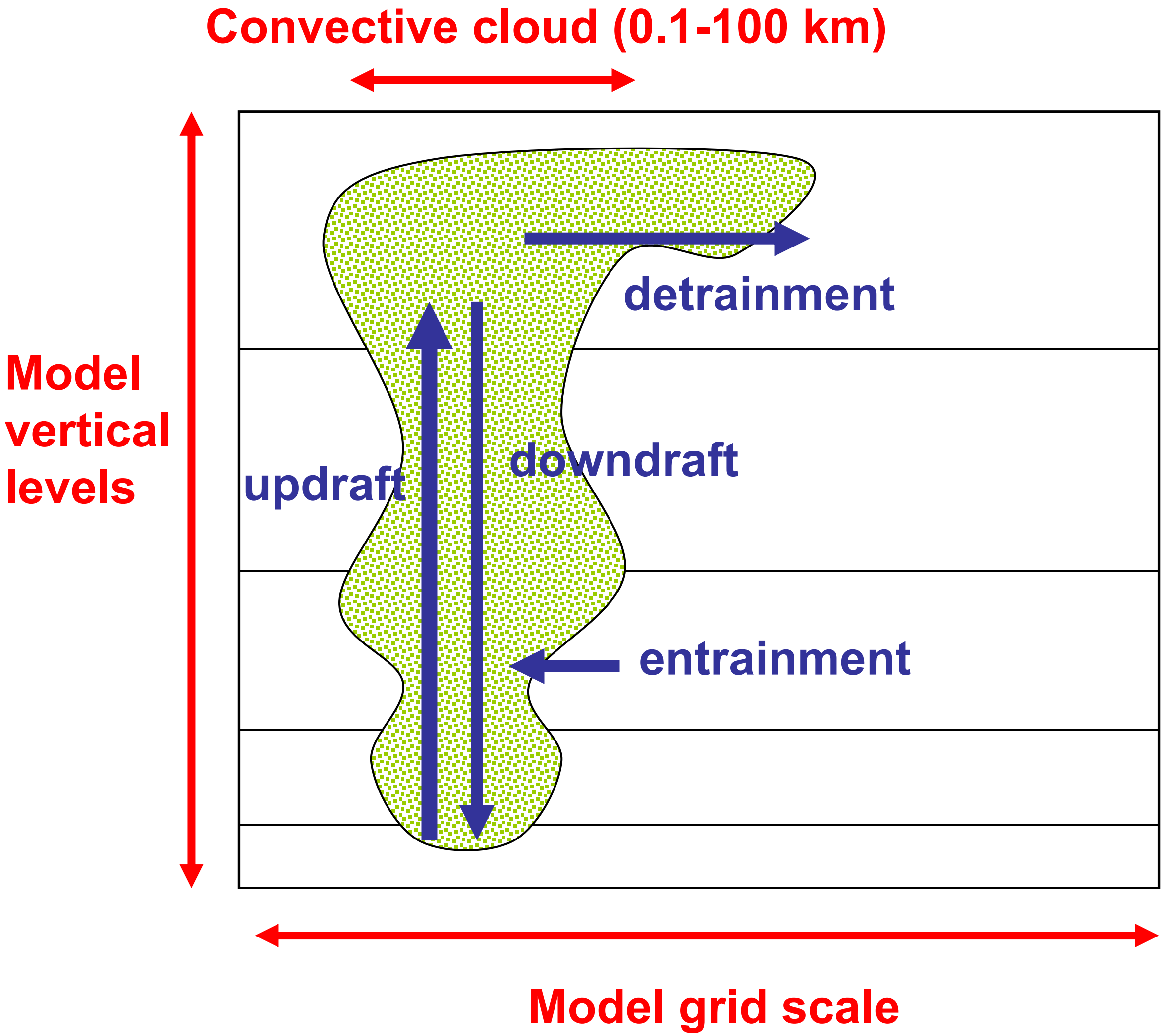
- Turbulent closure: eddy diffusivity
- Full tensor or simplifications (K-theory, etc.)
- Input from NWP (similarities with other eddies)
- Stability dependence
- Horisontal diffusion (depend on resolution)
- Vertical diffusion
- PBL height /Mixing height
- Other not included mechanisms

THE INFLUENCE OF ATMOSPHERIC STABILITY ON POLLUTION DISPERSION



Vertical turbulent transport (buoyancy)

- generally dominates over mean vertical advection
- K-diffusion OK for dry convection in boundary layer (small eddies)
- Deeper (wet) convection requires non-local convective parameterization



Wet convection is subgrid scale in global models and must be treated as a vertical mass exchange separate from transport by grid-scale winds.

Need info on convective mass fluxes from the model meteorological driver.

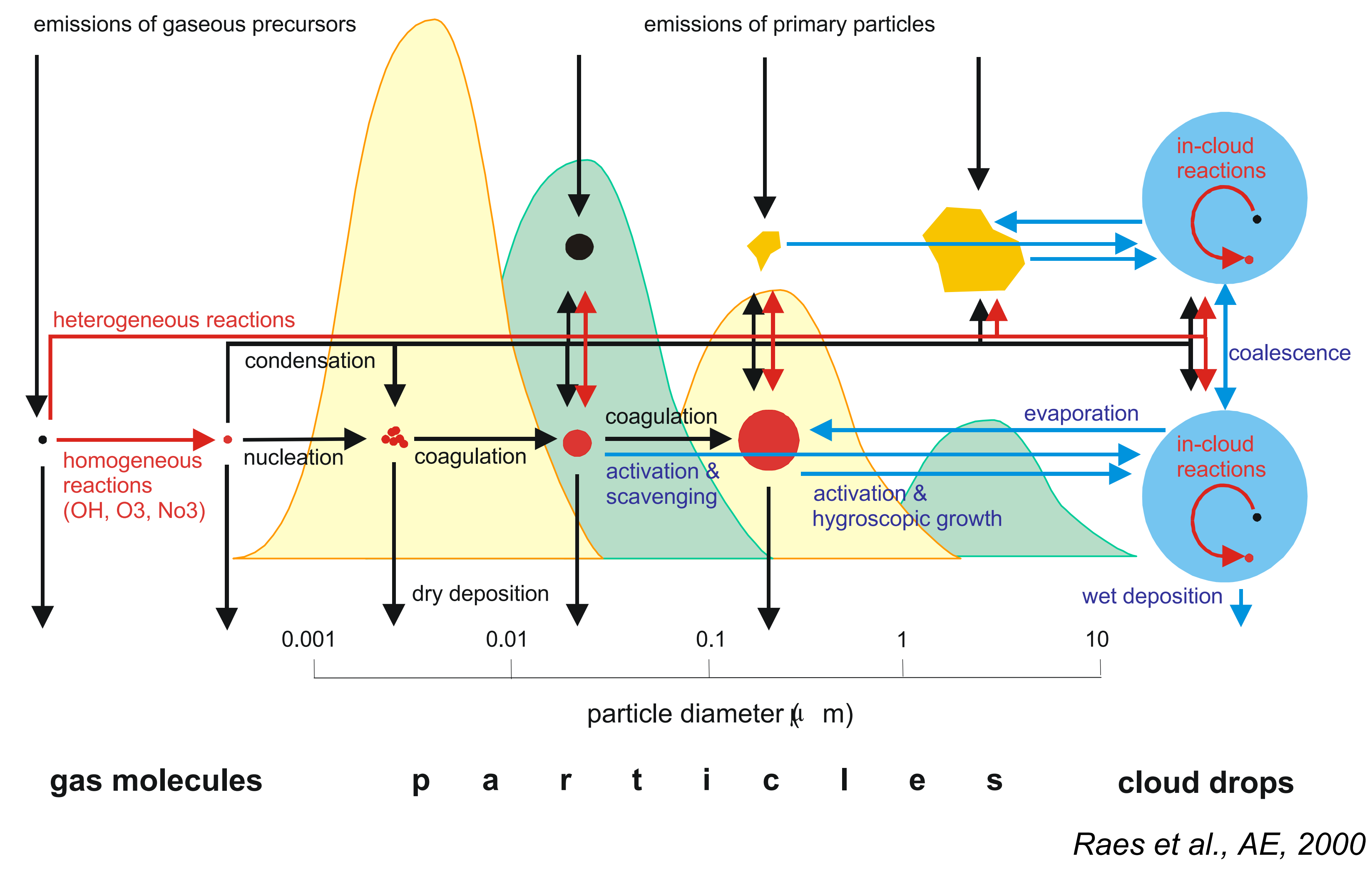
Inverse cascades and nonlocal mixing due to large-scale, organized eddies in the shear-free convection need to be included.

Chemical scheme

- One of the most time-consuming modules
- Contains of the most severe non-linearities, also the stiffest sub-system (several orders of magnitude of reaction time scales)
- Chemical kinetics

$$A + B \rightarrow C : \quad \frac{d[C]}{dt} = K[A][B]$$

Aerosol-CCN/IN dynamics modelling



$$\frac{dN(v,t)}{dt} = \left(\frac{dN(v,t)}{dt}\right)_{nucl} + \left(\frac{dN(v,t)}{dt}\right)_{cond} + \left(\frac{dN(v,t)}{dt}\right)_{coag} + \underset{(source)}{S} - \underset{(removal)}{R}$$

Dry and wet deposition

- Dry deposition
  - linear (well, sometimes)
  - surface process
  - moderate intensity
  - can be bi-directional (evaporation $\Rightarrow$ re-emission)
  - approached via e.g. resistive analogy (Wessely, 1989)
    - aerodynamic resistance
    - laminar-layer resistance
    - surface resistances: soil, canopy, water surface, …
    - sedimentation
  - detailed landuse needed

- Wet deposition
  - can be non-linear volume process
  - high intensity
  - high complexity and dependence on precipitation and species features =>
    - usually treated via "empirical" 1st-order equation:

$$\frac{\partial \varphi}{\partial t} = \Lambda(I, \ldots, \varphi)\varphi$$

  - where I is a precipitation intensity

Deposition mechanisms for aerosols

- Particle size dependend parameterisations for dry and wet deposition,
- Resistance approach for dry deposition,
- Terminal settling velocity in different regimes:
  - Stockes low,
  - non-stacionary turbulence regime,
  - correction for small particles,
- Different scavenging of particles and gases,
- Depending on classification of land/sea surface,
- Below-cloud scavenging (washout)
- Rainout between the cloud base & top (scavenging into cloud):
  - *convective* precipitation,
  - *stratiform* precipitation,
- Scavenging by snow.
- 3D cloud water and humidity available for deposition simulation

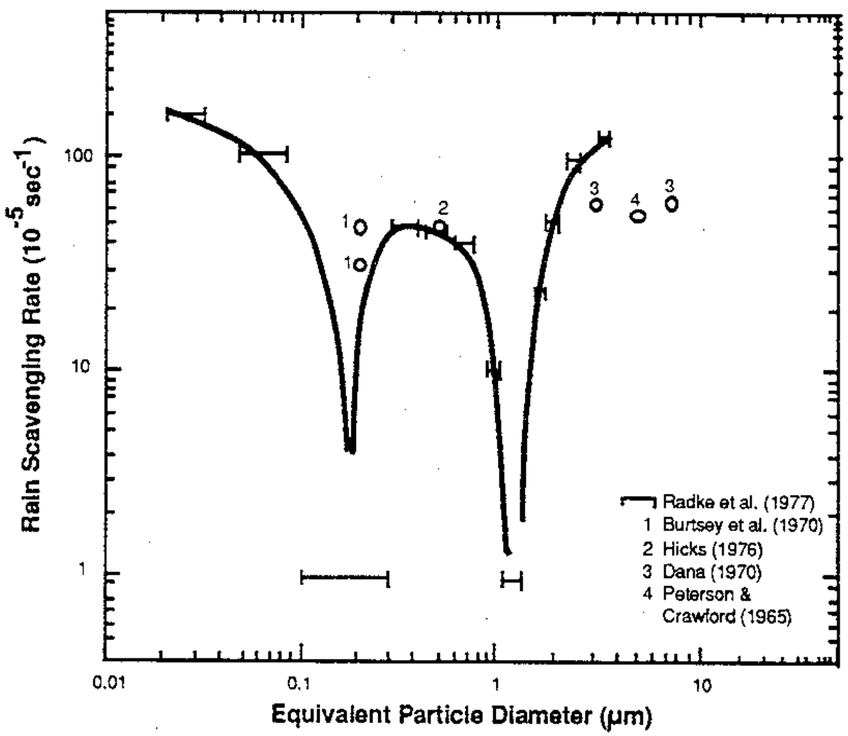
Wet deposition processes

- Below-cloud scavenging (washout) coefficient for aerosol particles of radius *rp*

$$\Lambda = -\pi N_r \int a^2 w_r(a) E(r_p,a) f_a(a)\,da,$$

- the 'Greenfield gap',

- Rainout between the cloud base & top (scavenging into the cloud):
  - *convective* precipitation,
  - *stratiform* precipitation,
- Scavenging by snow,
- Orographic effects (seeder-feeder effect),
- Deposition caused by surface fog.



Measured Values of Scavenging Coefficient vs. Particle Size (McMahon and Dennison, 1979))

Two formulations for the washout coefficient, Λ' (s-1)

1) as empirical function of particle radius r (μm) & rainrate q (mm/h):

$\Lambda'(r,q) = a_0\, q^{0.79}$, $\quad r < 1.4\ \mu m$

$\Lambda'(r,q) = (b_0 + b_1 r + b_2 r^2 + b_3 r^3)\, f(q)$, $\quad 1.4\ \mu m < r < 10\ \mu m$

$\Lambda'(r,q) = f(q)$, $\quad r > 10\ \mu m$

where $f(q) = a_1 q + a_2 q^2$, $a_0 = 8.4\cdot10^{-5}$, $a_1 = 2.7\cdot10^{-4}$, $a_2 = -3.618\cdot10^{-6}$, $b_0 = -0.1483$, $b_1 = 0.3220133$, $b_2 = -3.0062\cdot10^{-2}$, and $b_3 = 9.34458\cdot10^{-4}$.

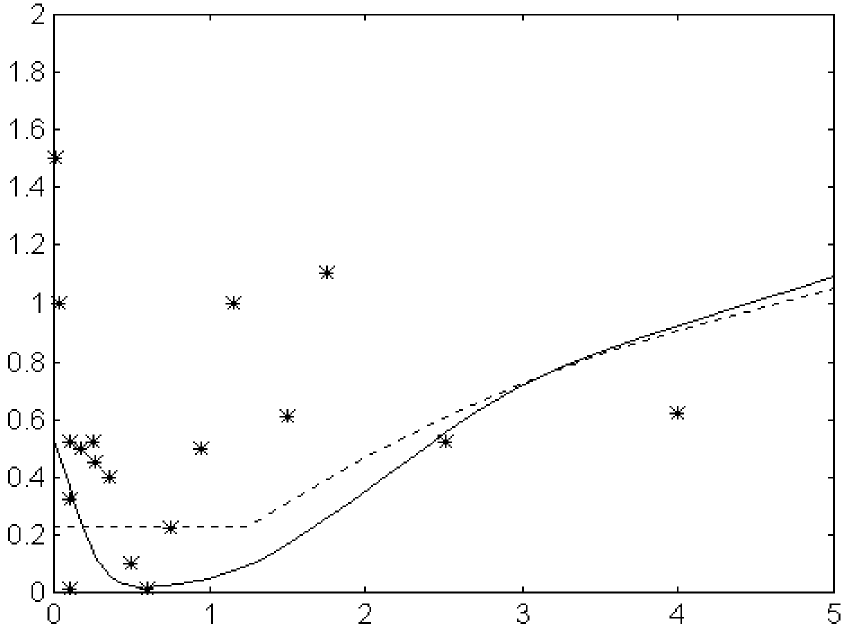
2) theoretical formulae for the Brownian capture mechanism, the aerosol capture efficiency due to the impaction of aerosol particles on the rain drop and interception of particles by the rain drop:

$$\Lambda = \frac{q}{2a_m}\left[\frac{4}{Pe}\left(1 + 0.4\,\mathrm{Re}^{1/2}\,Sc^{1/3}\right) + \frac{4r_p}{a_m}\left(\frac{r_p}{a_m} + \frac{(1 + 2\,\mu_w r_p/\mu_a a_m)}{\left(1 + \mathrm{Re}^{-1/2}\,\mu_w/\mu_a\right)}\right) + \left(\frac{\rho_w}{\rho_a}\right)^{\frac{1}{2}}\left(\frac{St - St_*}{St - St_* + 2/3}\right)^{\frac{3}{2}}\right]$$

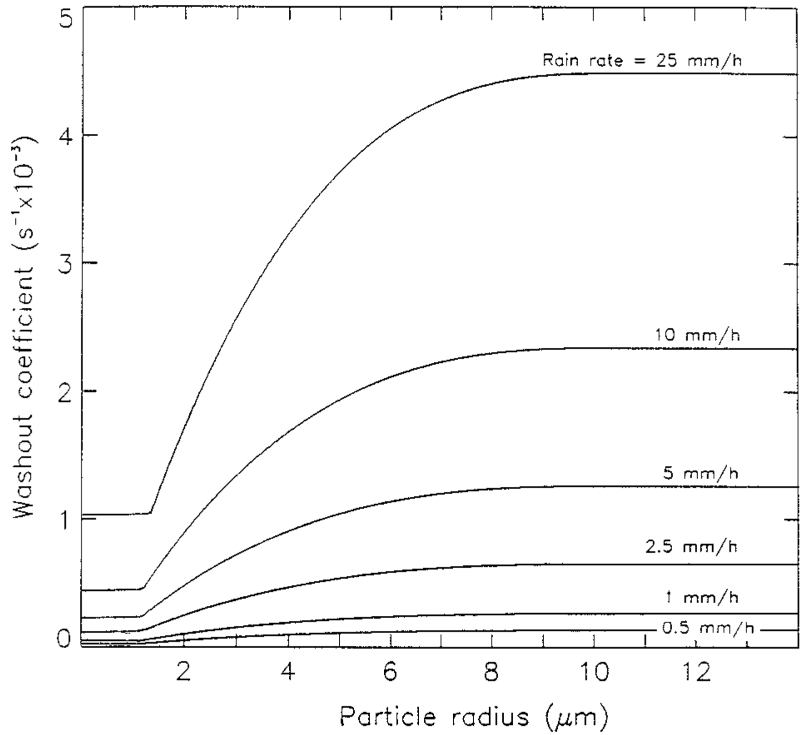
where a_m is the volume-mean raindrop projected radius, St - the Stokes number $(-2r_p^2\rho_p w_r/9$ ♋ $\rho_a \nu)$, St_* - the critical Stokes number $(1.2+\ln(1+Re)/12)/(1+\ln(1+Re))$, μ_w and μ_a - the dynamic viscosities of water and air, respectively, and ρ_p, ρ_w and ρ_a - the density of particles, water and air, respectively, Pe - the Peclet number (aw_r/D), Sc -the Schmidt number (ν/D), Re - the Reynolds number (aw_r/ν), ν - the kinematic viscosity of the air (μ_a/ρ_a), and D - the Brownian diffusivity of particles.

Baklanov and Sørensen, 2001

Wet deposition processes



Dependence of the washout coefficient on particle radius for a rain intensity of 5 mm/h.



Dependence of the washout coefficient on particle radius and rain intensity.

Baklanov and Sørensen, 2001

Diagnostic and output post-processing

- Computation of diagnostic variables
  - e.g. optical features of the atmosphere from concentrations
  - proxies for health impact and risk assessment
- Transformation from model-convenient variables to user-friendly ones
  - generation of integrated / averaged variables
- Conversion to convenient file formats
- Grid interpolation (if needed)

Main components of model evaluation

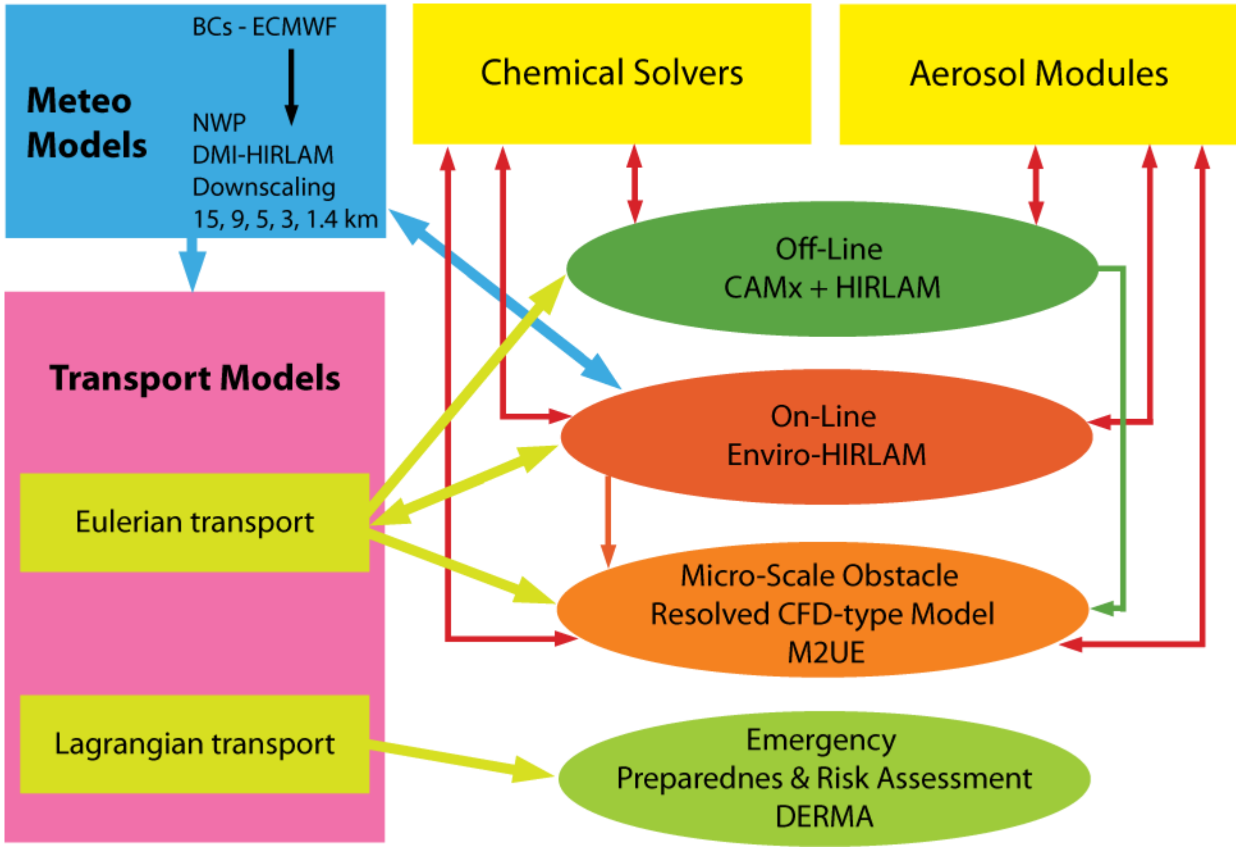
1. **Operational Model Evaluation** involves the direct comparison of model output with analogous observations in an overall sense. It utilizes routine observations of ambient pollutant concentrations, emissions, meteorology, and other relevant variables.
2. **Diagnostic Model Evaluation** examines the ability of a model to predict pollutant concentrations by correctly capturing physical and chemical processes, and their relative importance as incorporated in the model. This type of model evaluation generally requires detailed atmospheric measurements that are not routinely available.
3. **Dynamic Model Evaluation** focuses on model's ability to predict changes in air quality levels in response to changes in either source emissions or meteorological conditions. This exercise requires historical case studies where known emission changes or meteorological changes occurred that could be confidently estimated.
4. **Probabilistic Model Evaluation** attempts to capture statistical properties, including uncertainty or level of confidence in the model results for air quality management or forecasting applications; this approach is based on knowledge of uncertainty imbedded in both model predictions and observations.

Model evaluation: A few thoughts

- Careful selection of evaluation data sets necessary (representativeness, measurement biases, etc.)
- Advantages and disadvantages of satellite versus in-situ data
- What parameters most important to test integrated models?
  - Chemistry
  - Emissions
  - Deposition
  - Direct aerosol effects
  - Indirect aerosol effects
- Time scales for evaluation very important:
  - statistically significant results may only be obtained for long integration periods
- Practical issues
  - availability of data sets
  - data formats
  - model output for evaluation (what time resolution needed?)

DMI Multi-Scale MetM and ACT Modelling System

Baklanov, Mahura, Sokhi (Eds), ***2011****:*
"Integrated Systems of Meso-Meteorological and Chemical Transport Models", Springer, 242p.



Regional (European) scale: smog and ozone forecast and impact assessments.

Regional (European) to urban scale: smog and ozone, pollen forecast, two-way feedbacks
Climate version: EnvClim

Street scale

Nuclear, veterinary and chemical