JÜLICH
FORSCHUNGSZENTRUM

Numerical Simulations of Atmospheric Compounds

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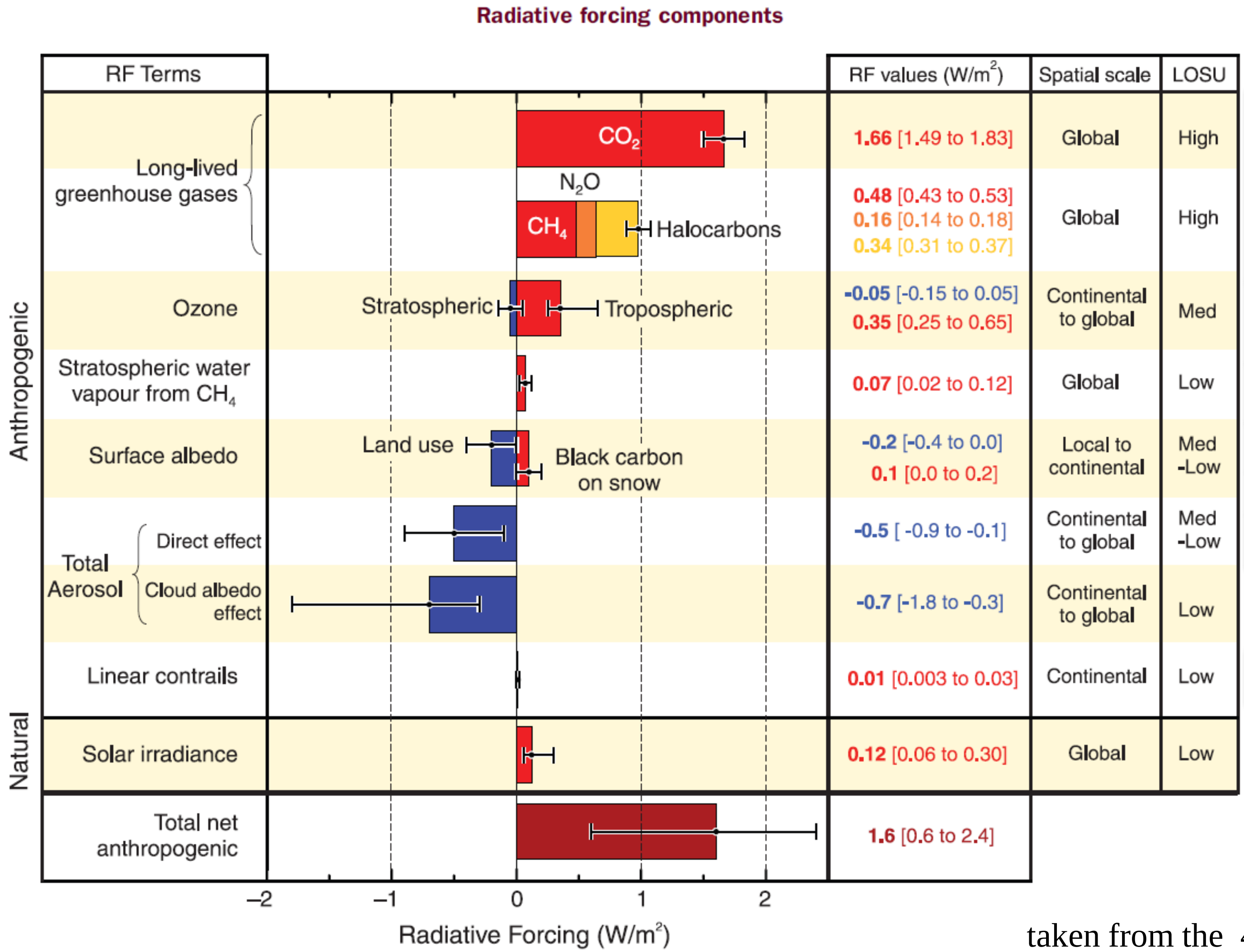
Outline

1. Objectives: Why atmospheric chemistry modelling? Two reasons:
   1. for climate modelling
   2. for air quality modelling
2. How can we model atmospheric chemistry?
   the advection (convection)-diffusion-reaction equation
3. Examples
   1. regular nested forecasts
   2. field mission support

1. Why atmospheric chemistry for climate modelling?
Uncertainties on climate radiative forcing is mainly induced by chemical components



dir. chem.

dir. chem.

dir. chem.

no chem.

indir. chem.

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no chem.

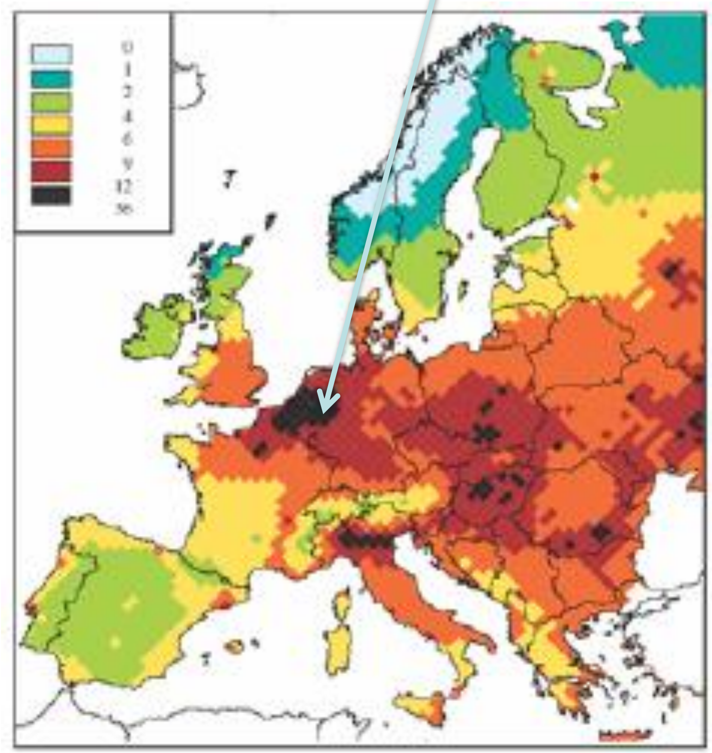
taken from the 4th IPCC Assessment Report,

1. Why atmospheric chemistry for climate modelling?
“Aerosol climate“ affects health.

The impact of PM_x (Particulate Matter) on human health can be severe.
CAFE (Clean Air For Europe, 2005) final report states:

- Estimated reduction of lifetime expectance due to $PM_{2.5}$ is large
- Modelling of TPM is not yet satisfying
  - *Too few measurements*
  - *Too large model uncertainties*

How to
get to a more reliable estimation of aerosol load and exposure?
- react on sudden emission changes?
- deal with heterogeneous data?



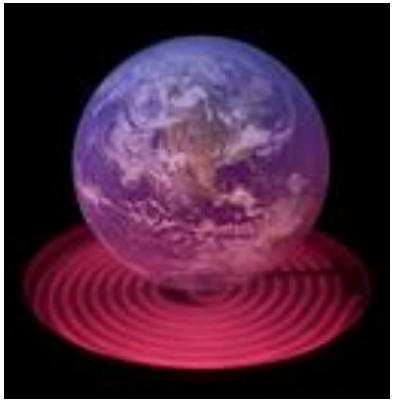
Estimated $PM_{2.5}$ caused reduction of lifetime expectance in months (Emissions 2000)[CAFE]

A major effort in EU: GEMS→MACC→MACC-II an integrated approach for the atmospheric climate component www.gmes-atmosphere.eu/

Monitoring Atmospheric Composition and Climate - Interim Implementation - provides **data records on atmospheric composition** for recent years, data for **monitoring present conditions** and **forecasts of the distribution of key constituents** for a few days ahead. MACC-II combines state-of-the-art atmospheric modelling with Earth observation data to provide information services covering

European Air Quality

Global Atmospheric Composition

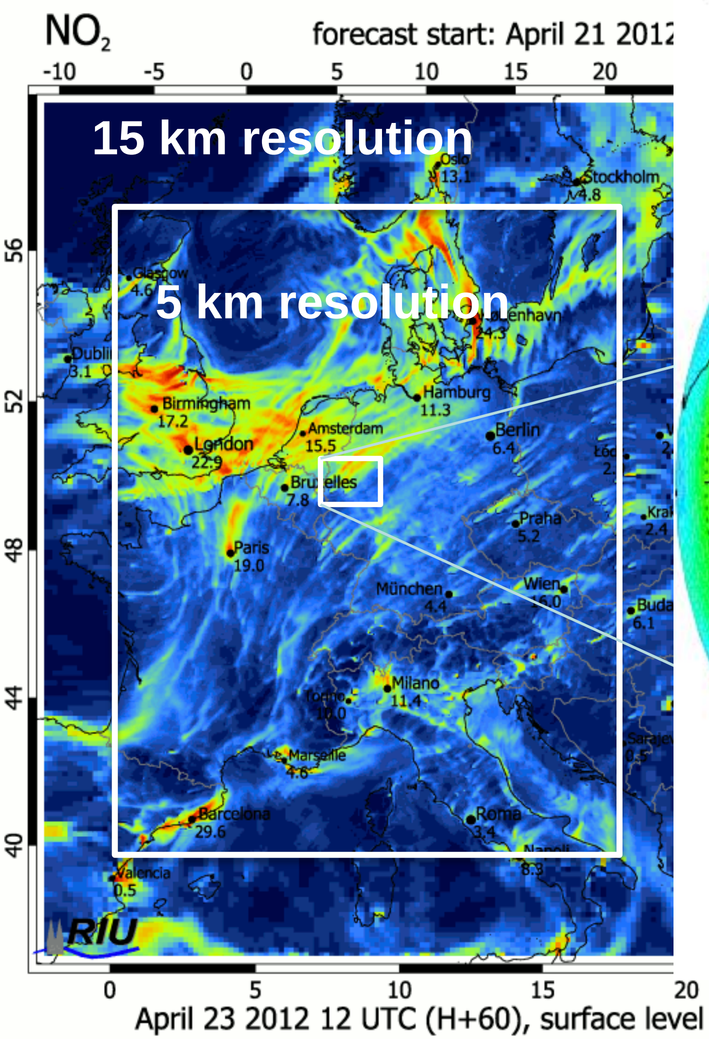


Climate

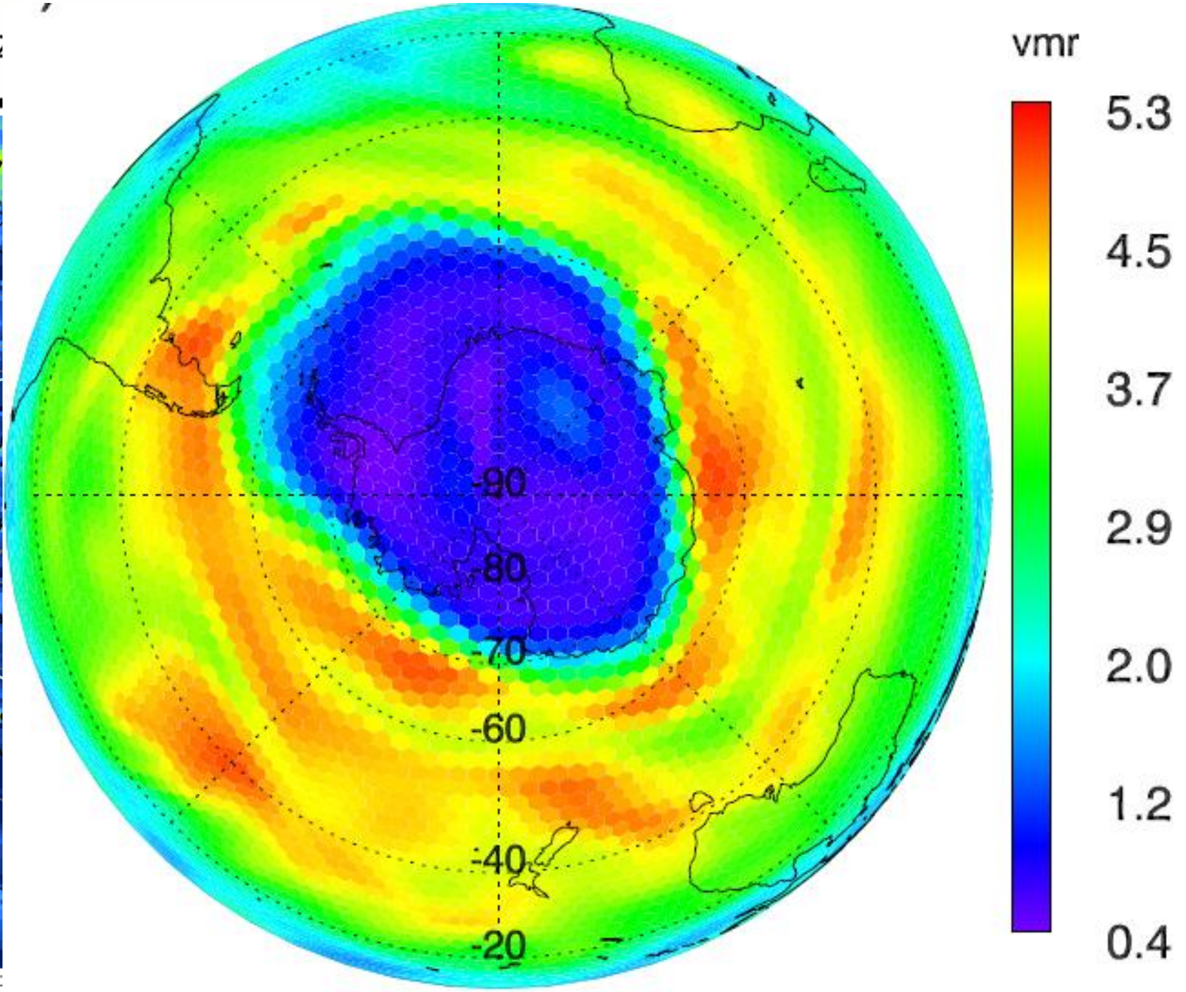
UV, solar energy

Spacial scales: from global to local

EURAD-IM nesting



SACADA southern polar vortex



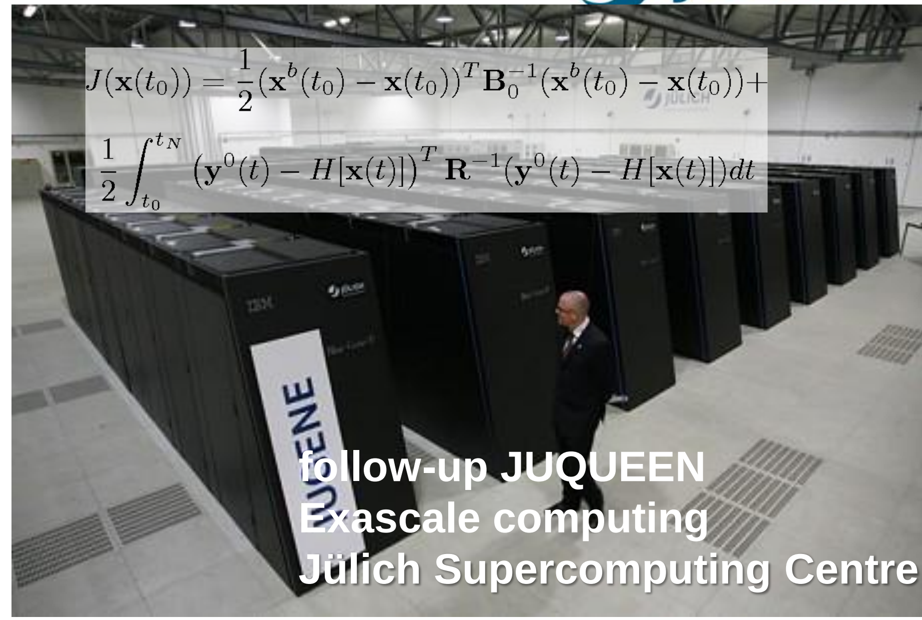
ozone at 44 hPa, 18.8.2002

GMES (Global Monitoring for Environment and Security)

How do we learn most of the climate state?

How can we enlist all information sources for my very special focus?

Hardware for models



Hardware for data

A close relation to “Earth Observation”

„Earth Observation“ comprises scientific activities, aiming at

- exploring of environmental factors, which harm human health,
- exploring climate evolution and its driving forces,
- mitigation of losses from disasters

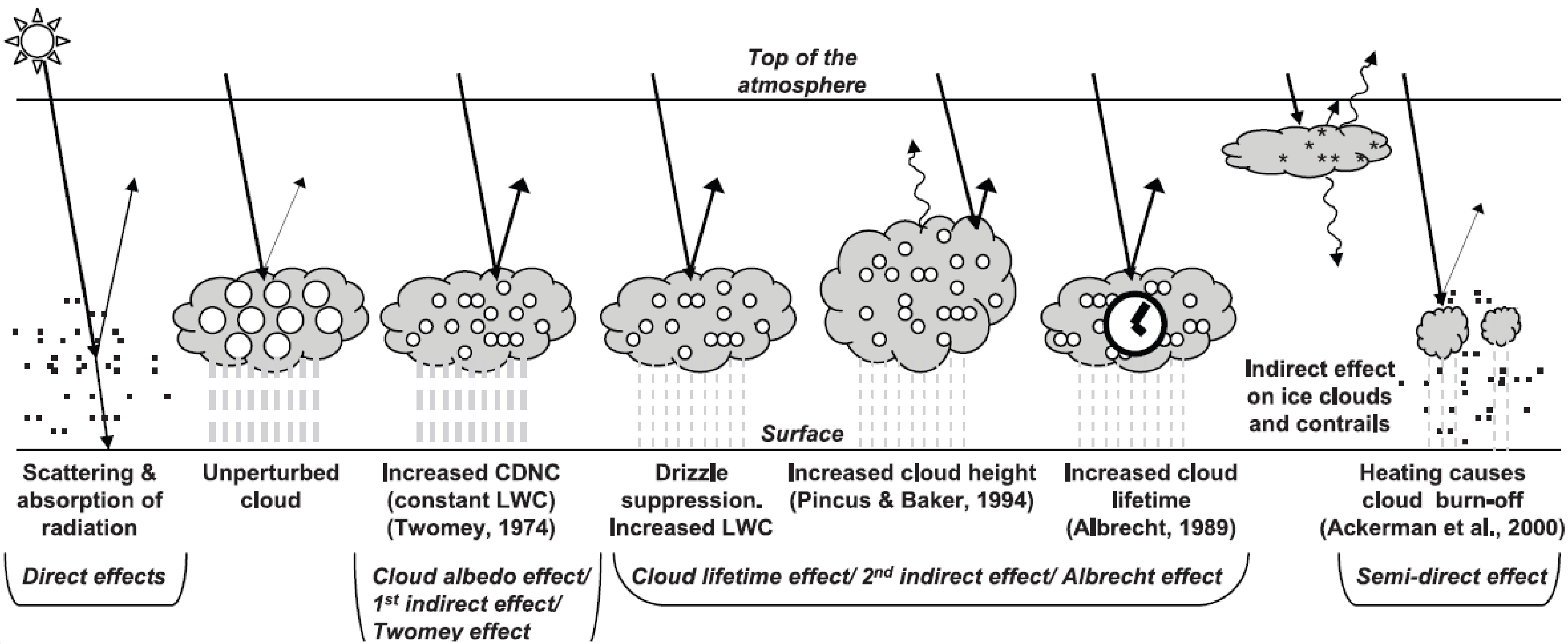
A key objective: Estimate emissions!!

→Use Data Assimilation and Inverse Modelling

<u>Global:</u> "Group on Earth Observations" (GEO) and :„Global Earth Observing System of Systems“ (GEOSS)

<u>European</u>:
Global Monitoring for Environment and Security (GMES)
Projekte MACC, PASODOBLE,…

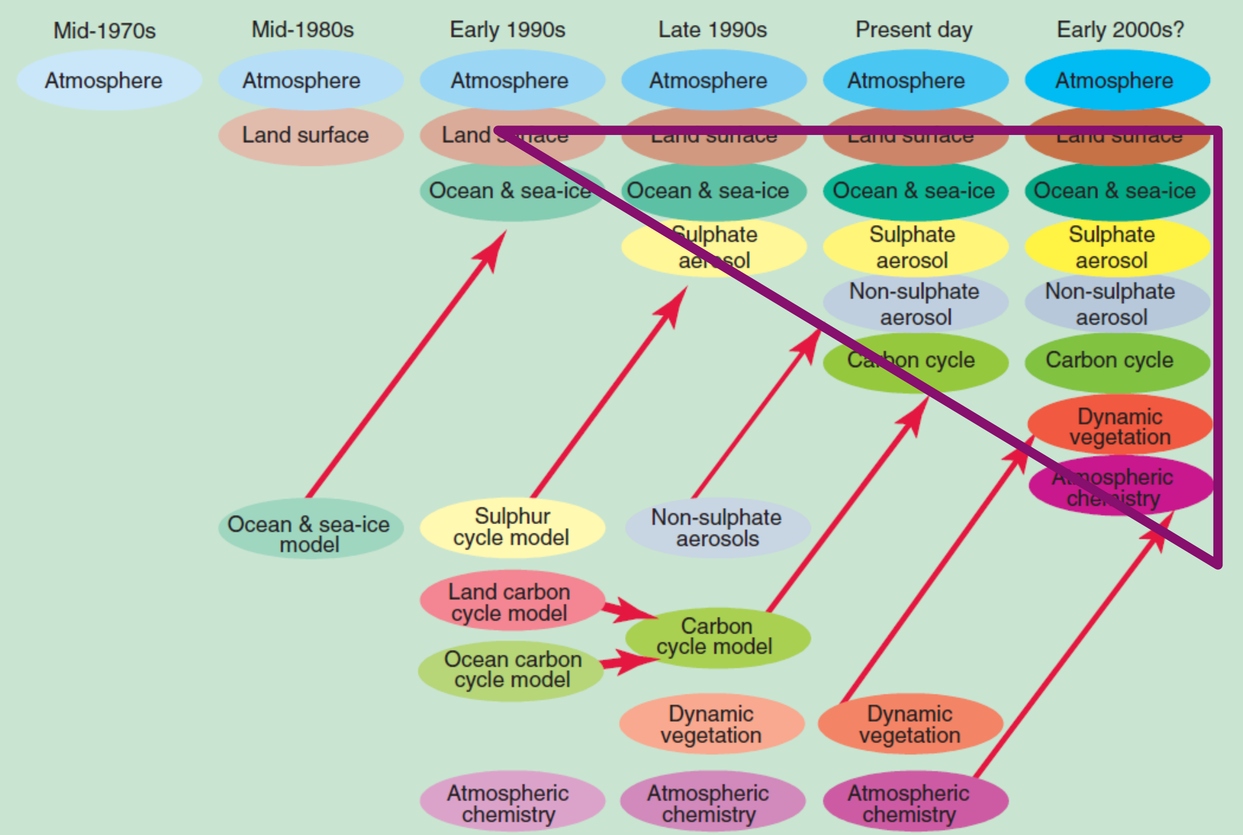
1. Why atmospheric chemistry for climate modelling?
Again, aerosols act via manifols pathways.



IPCC AR4,
modified from Haywood and Boucher, 2000

2. How can we model atmospheric chemistry?
Most of recent additions concern atmospheric chemistry

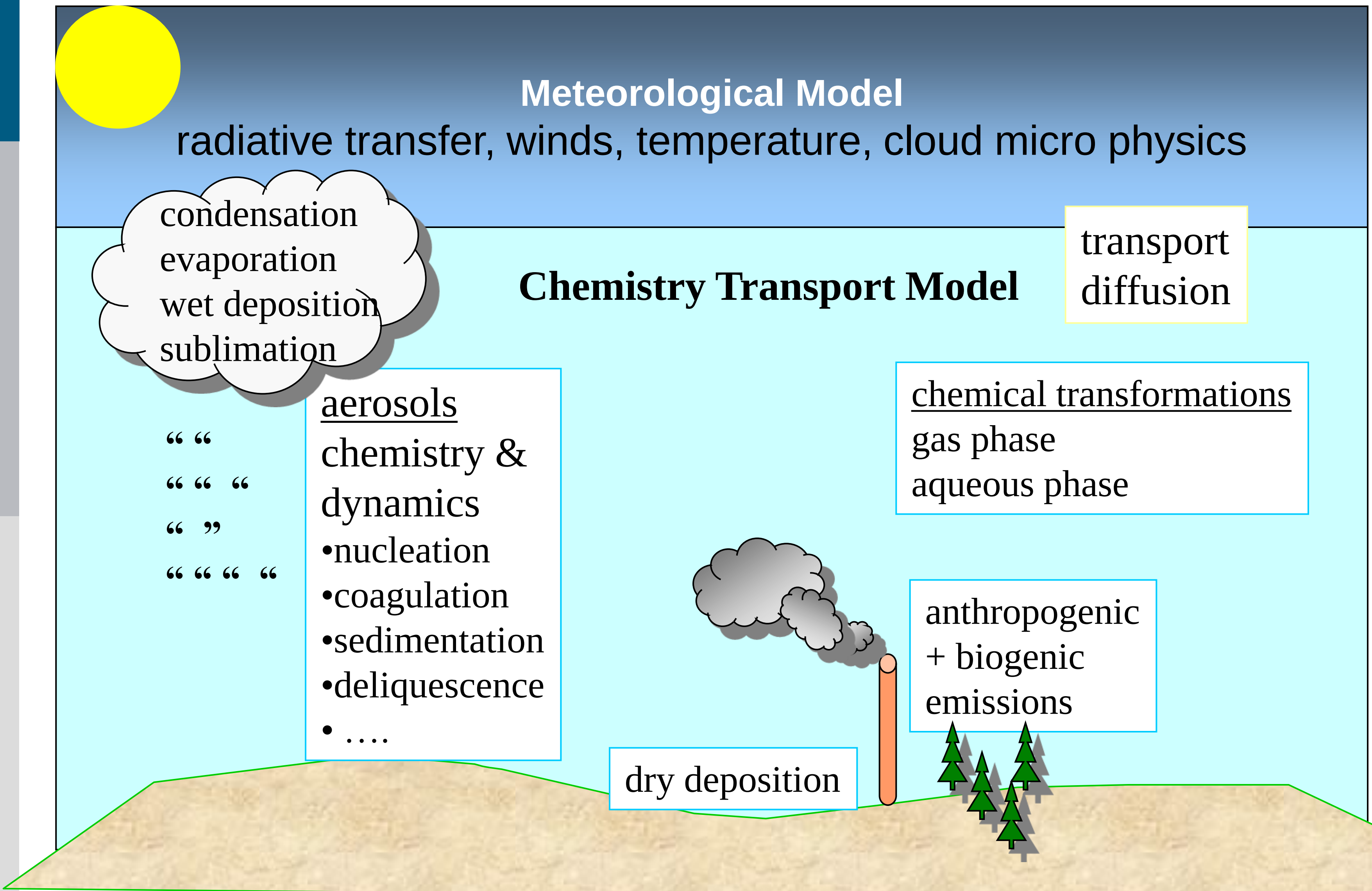
The Development of Climate models, Past, Present and Future



Box 3, Figure 1: The development of climate models over the last 25 years showing how the different components are first developed separately and later coupled into comprehensive climate models.

IPCC AR4

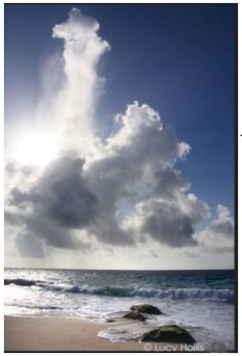
2. How can we model atmospheric chemistry?
Processes in a complex chemistry-transport model



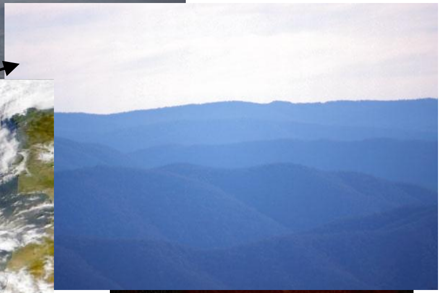
Scope of Simulations: TroposphereChemistry-Transport model EURAD-IM
(Reaction-Advection-Diffusion equation)
and further simulated processes

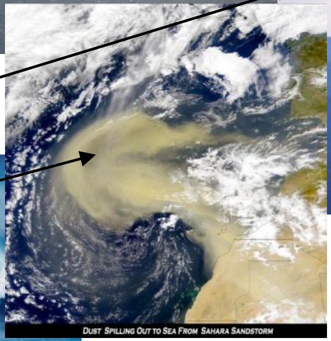
Chemistry mechanisms,

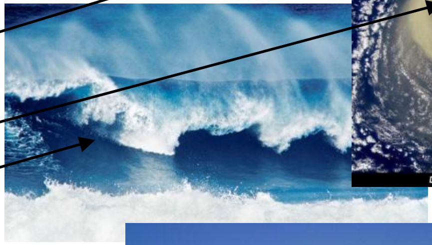
60 Gas phase constituents



Aqueous phase chemistry



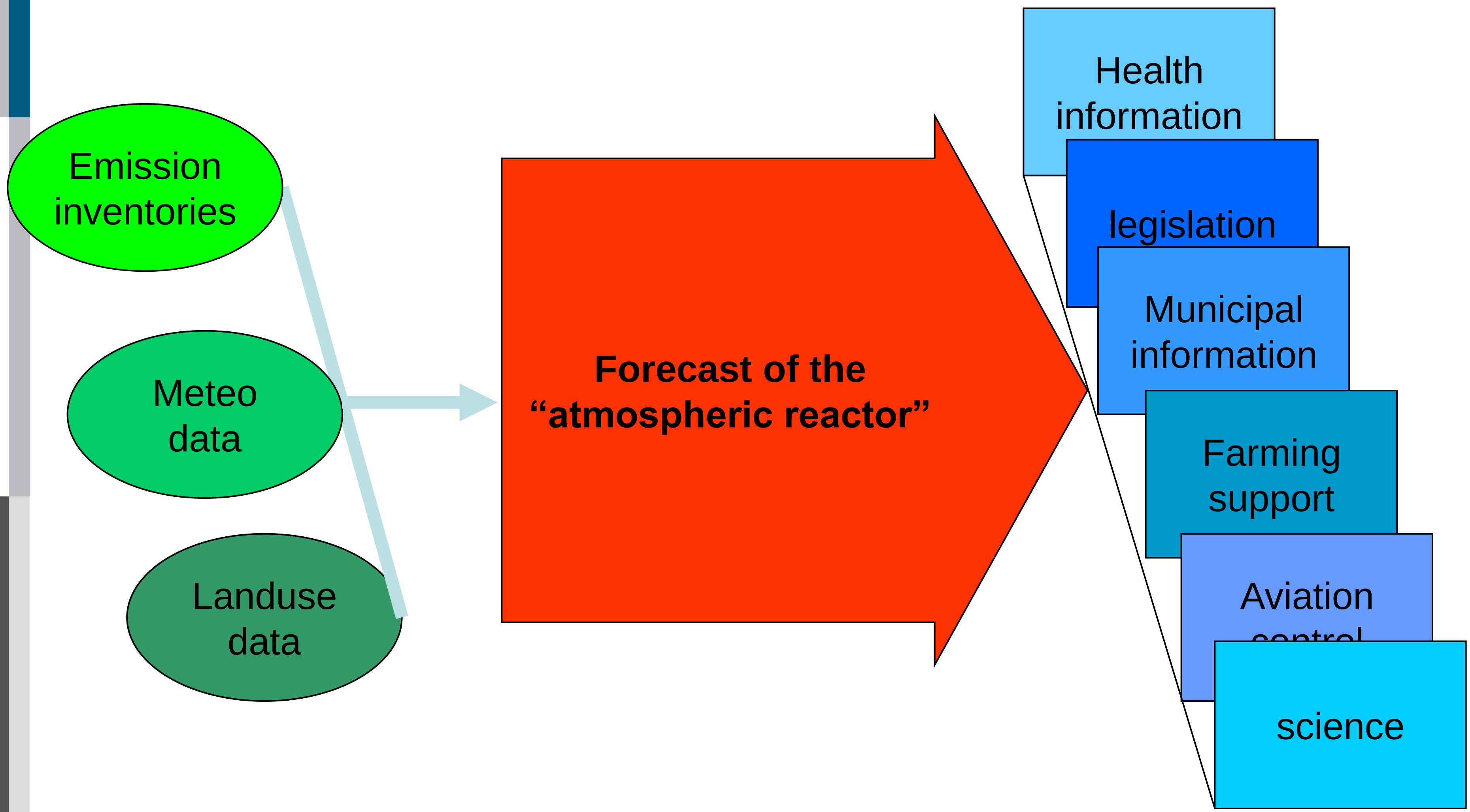




Aerosols

- anorganic
- secondary organic
- mineral dust
- sea salt
- biomass burning
- vulcanic emissions

"Upgrade data to information"



Transport-diffusion-reaction equation

Tendency Equations

direct chemistry transport equation

$$\boxed{\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{v} c_i) - \nabla \cdot (\rho \mathbf{K} \nabla \frac{c_i}{\rho}) - \Sigma_{r=1}^{R} \left(k(r)\,(s_i(r_+) - s_i(r_-))\,\Pi_{j=1}^{U} c_j^{s_j(r_-)} \right) = E_i + D_i}$$

c_i	concentration of species i	c_i^*	adjoint of concentration of species i
$\mathbf{v}$	wind velocity	s	stoichiometric coefficient
$k(r)$	reaction rate of reaction r	$\mathbf{K}$	diffusion coefficient
U	number of species in the mechanism	R	number of reactions in the mechanism
E_i	emission rate of species i (source)	D_i	deposition rate of species i (sink)

adjoint chemistry transport equation

Chemistry processes extremely diverse:
- gas phase reactions (ODEs as given above)
- aqueous phase (well mixed solution in droplets)
- aerosols (surface reactions)

Chemistry and transport models (CTM)

off-line models

– First determine the meteorological fields solving the equations of motion (Navier[1827]-Stokes[1845]-equations),...

– Then solve the balance equations for trace compounds

– no feedback of the chemistry on transport

• full coupling: simultaneous determination of the meteorological fields and the trace gas budgets

– feedback of the chemistry on the dynamics (meteorology) via the physical properties of trace compounds [important for climate modelling]

Models in atmospheric chemistry

Based on a quantitative understanding of the physical and chemical processes equations are developed that can be used to calculate the temporal evolution and the spatial distributions of trace gases.

– theory: equations of motion and the continuity equations from fluid dynamics, state

equation of gases,...

– phenomenological models: for instance dry deposition in analogy to electric circuits

Balance equations I

Relation between the state of a system and the change of the state.

Examples:

- Radioactive decay

$$\frac{dN(t)}{dt} = -1/\tau N(t)$$

- Newton's equation of motion

$$m\frac{d^2x(t)}{dt^2} = K(x(t))$$

- response X of the immune system on an attack by pathogenic agents P(r)

$$\frac{dX(t)}{dt} = +\kappa \int_{body} P(r)dr$$

Additivity

Consider the first order decay of X via processes 1 and 2

$$
\begin{aligned}
\left(\frac{d[X]}{dt}\right)_1 &= -k_1[X] \\
\left(\frac{d[X]}{dt}\right)_2 &= -k_2[X] \\
[X]_1 &= [X(0)]\exp(-k_1 t) \\
[X]_2 &= [X(0)]\exp(-k_2 t)
\end{aligned}
$$

Additivity of the balance equations

$$\left(\frac{d[X]}{dt}\right) = \left(\frac{d[X]}{dt}\right)_1 + \left(\frac{d[X]}{dt}\right)_2$$

$$[X(t)] = [X(0)]\exp(-(k_1 + k_2)t) \neq [X(t)]_1 + [X(t)]_2$$

Balance equation II

Coupled partial differential equations in space and time

rate of chemical production P_i and the frequency of destruction D_i are in general non

linear functions of concentrations

$$\mathbf{C} = (C_1(\mathbf{r}, t), \ldots, C_n(\mathbf{r}, t))$$

$$\frac{\partial C_i(\mathbf{r}, t)}{\partial t} = P_i(\mathbf{C}) - D_i(\mathbf{C})C_i + T(C_i)$$

Numerical aspects (Chemistry)

The chemical balance equations (nonlinear ordinary 1. order differential equations) are stiff because of the large variability of relaxation times (life times) between nano seconds (O(1D)) and many years (CH4).

Standard numerical integration requires time steps which are much smaller than the smallest relaxation time.

- Gear's solver (often used as reference)
- applying the quasi-stationary state approximation (QSSA) to reduce stiffness. The balance (differential) equations for short lived compounds are replaced by algebraic equations $C_{QSSi} \approx P_i(C)/D_i(C)$

Transport I

Transport caused by *divergence* of the flux (more precisely: current density)

$$\mathrm{T}(\mathrm{C_i}) = -\nabla \bullet (\mathbf{v}\mathrm{C_i})$$

in cartesian coordinates

$$\mathbf{v} = (\mathrm{u}, \mathrm{v}, \mathrm{w}) \qquad \mathbf{r} = (\mathrm{x}, \mathrm{y}, \mathrm{z})$$

$$\mathrm{T}(\mathrm{C_i}) = -\frac{\partial(\mathrm{uC_i})}{\partial \mathrm{x}} - \frac{\partial(\mathrm{vC_i})}{\partial \mathrm{y}} - \frac{\partial(\mathrm{wC_i})}{\partial \mathrm{z}}$$

The transport $\mathrm{T}(\mathrm{C_i})$ is a linear operator.

Purpose of box-models

study of chemistry and identification of key compounds and processes

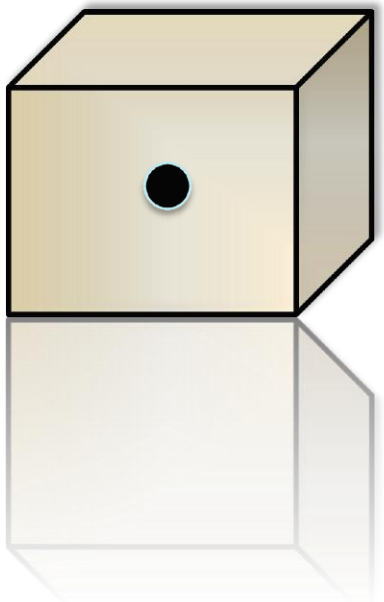
chemical life time

separation of the influence of transport

validation of chemical reaction mechanisms

(observationally constrained modelling)

applied to chamber experiments



Balance equations III

Goal: Calculate Ci(r, t)

- the wind field v(r,t) and other meteorological fields have to be determined prior to the

CTM simulation (for example with a weather prediction model)

- input data (Initial conditions at t = 0 and boundary conditions for the whole simulation

period t = ...T are required):

measured data, from other model calculations, back ground data, ...

Influence depends on the scale of the model.

- Information on emission strengths and non chemical losses are required.

Only numerical solution possible (Quite a job!)

1-dimensional model

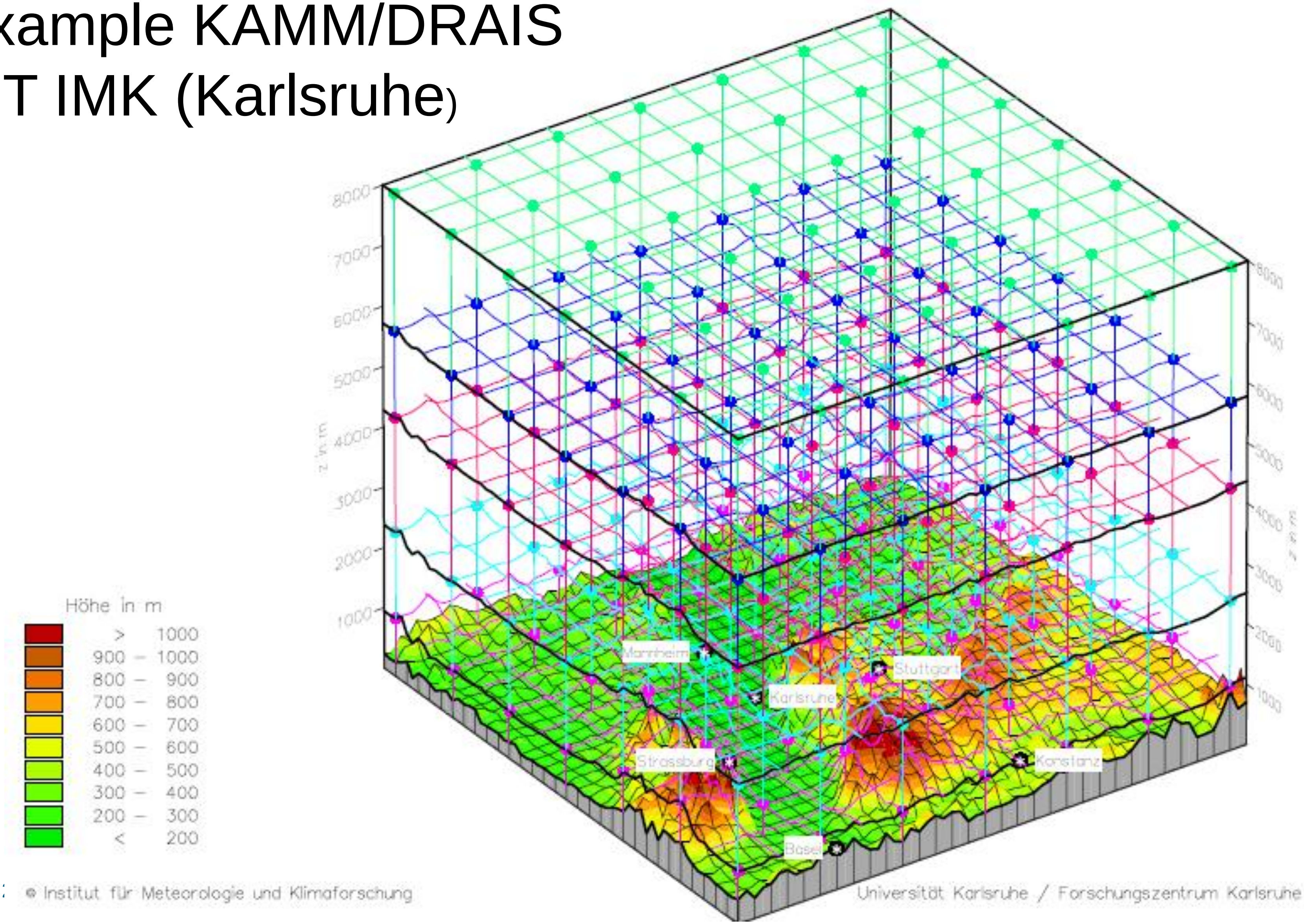
Dependence of concentrations on height z

assumption: horizontally well mixed

$$\frac{\partial \mathrm{C_i}(\mathrm{z}, \mathrm{t})}{\partial \mathrm{t}} = \mathrm{P_i}(\mathbf{C}) - \mathrm{D_i}(\mathbf{C})\mathrm{C_i} - \frac{\partial(\mathrm{wC_i})}{\partial \mathrm{z}}$$

To be solved under initial conditions C(z, t = 0) und boundary conditions for concentrations and fluxes (sources and sinks) at the upper and lower boundary of the model.

3-D models
Example KAMM/DRAIS
KIT IMK (Karlsruhe)



Discretisation of the 1-D advection equation (1)

The advection equation is a special case of
quasi-linear partial differential equation of 1. order

$$\boxed{\frac{\partial u(t,\mathbf{r})}{\partial t} + \mathbf{v}(t,\mathbf{r}) \cdot \nabla u(t,\mathbf{r}) = g(t,\mathbf{r})}$$

It describes transport of a material property u(t,**r**) advected by speed **v** with generation and destruction g(t,r) in spatio-temporal dependency (t,**r**).

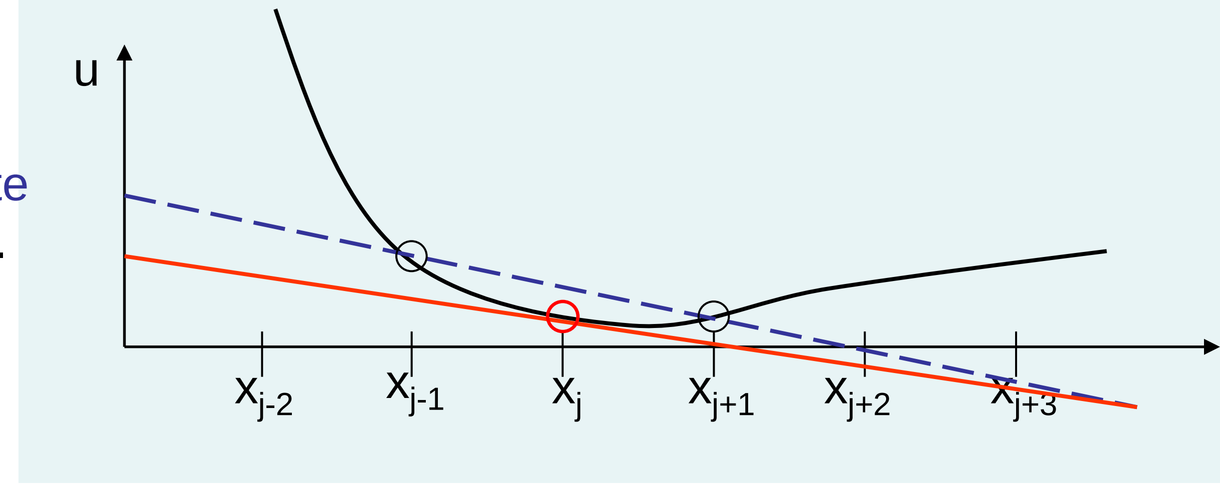
Discretisation of the 1-D advection equation (2)

spatial

One possible discretisation of the spatial derivative are "centred differences"

approximate

exact deriv.



$$\frac{\partial u}{\partial x} =: u_x$$

$$u_x|_{x_j} = \frac{u_{j+1} - u_{j-1}}{2h} + \frac{h^3}{12}\left(u_{xxx}|_{\xi_+} + u_{xxx}|_{\xi_-}\right)$$

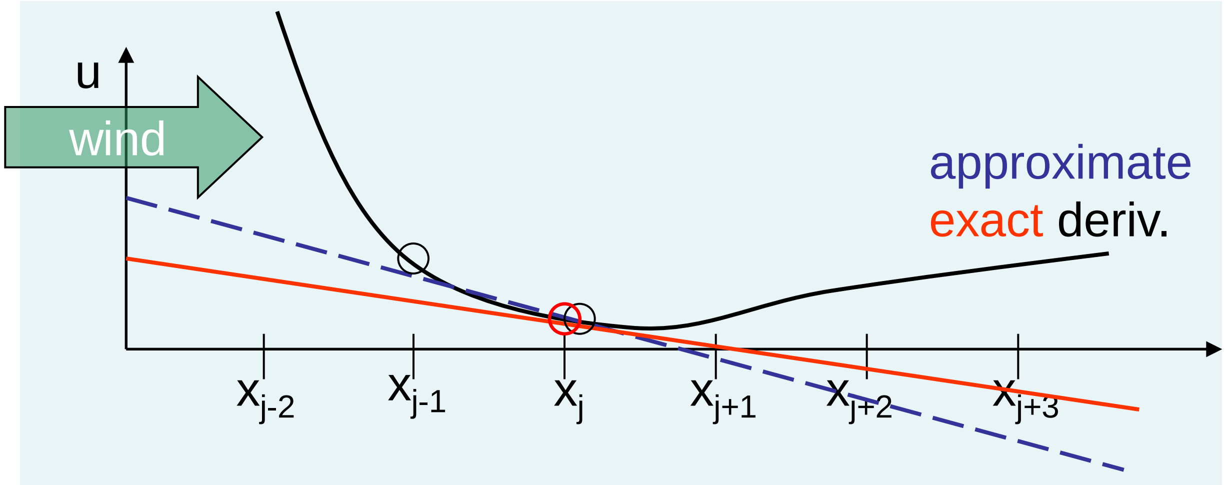
with $\xi_+ \in (x_j, x_{j+1})$ and $\xi_- \in (x_{j-1}, x_j)$, as following Taylor

$$u_{j\pm1} = u_j \pm h u_x|_{x_j} + \frac{h^2}{2} u_{xx}|_{x_j} \pm \frac{h^3}{6} u_{xxx}|_{\xi\pm}$$

approximation is of quadratic order $\mathcal{O}(h^2)$.

Discretisation of the 1-D advection equation (3) spatial

Another option is “upstream discretisation because the 2nd node is selected in windward direction (more predictive skill what to expect at node u_j.)



$$u_x|_{x_j} = \frac{u_j - u_{j-1}}{h} + \frac{h}{2} u_{xx}|_\xi, \qquad \xi \in (x_{j-1}, x_j)$$

Error is of linear order only!

Discretisation of the 1-D advection equation (3) temporal

The temporal discretisation like “upwind scheme”.

("forward in time"), with $t_n := n \cdot \Delta t, u_j^n = u(n \cdot \Delta t, j \cdot h)$, denn

$$u_t|_{tn} = \frac{u_j^{n+1} - u_j^n}{\Delta t} - \frac{\Delta t}{2} u_{tt}|_\eta, \qquad \eta \in (t_n, t_{n+1}).$$

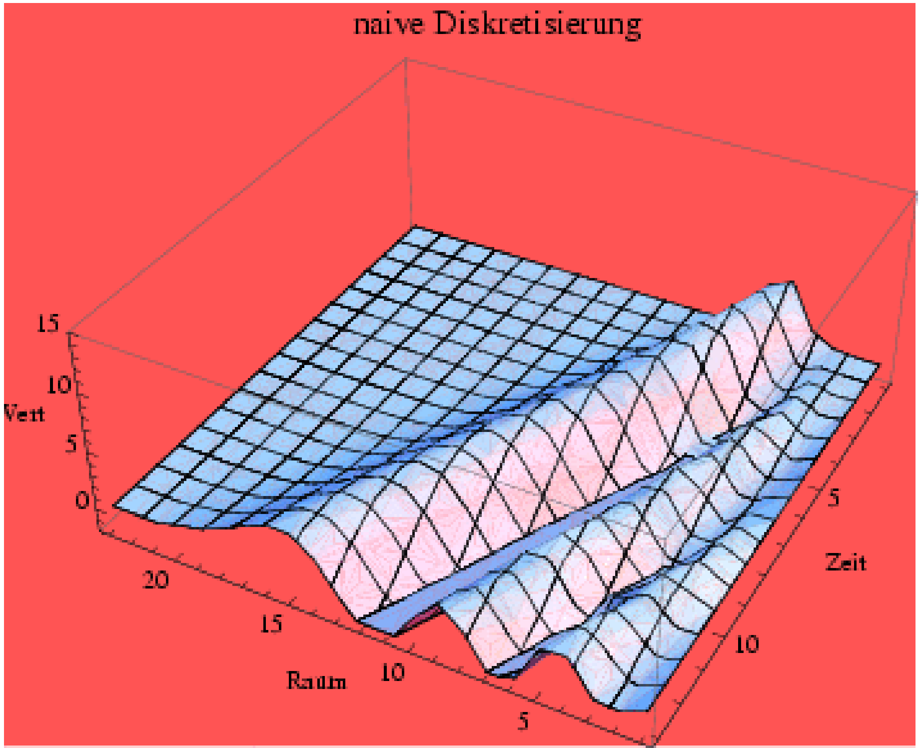
linear order $\mathcal{O}(\Delta t)$.

Discretisation of the 1-D advection equation (3) spatial

Alltogether, the 1-dim. discretized form of equation $\frac{\partial u(t,\mathbf{r})}{\partial t} + \mathbf{v}(t,\mathbf{r}) \cdot \nabla u(t,\mathbf{r}) = g(t,\mathbf{r})$

$$\frac{u_j^{n+1} - u_j^n}{\Delta t} + \frac{c}{2h}\left(u_{j+1}^n - u_{j-1}^n\right) = \tau_j^n,$$

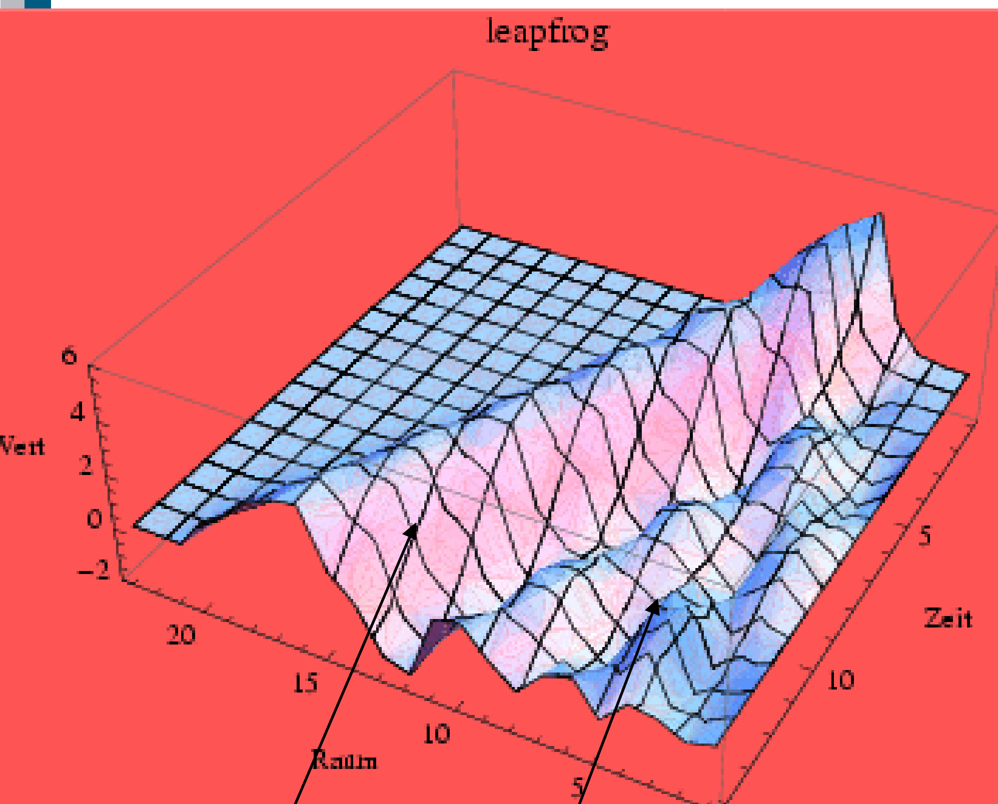
with local *truncation error* τ_j^n of order $O(\Delta t) + O(h^2)$.

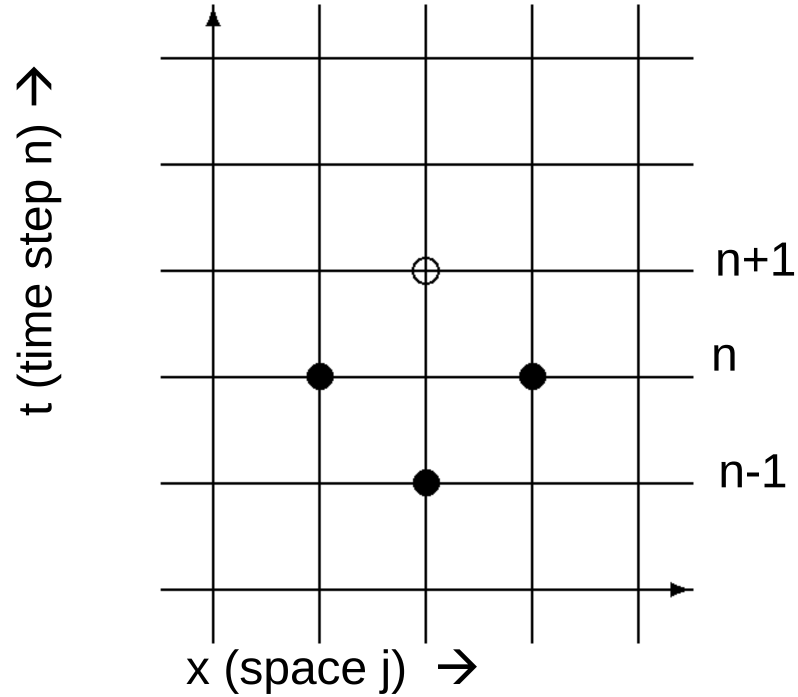


Not that what we can need!!

Leapfrog method for time integration

$\lambda = \frac{c \cdot \Delta t}{\Delta x} \leq 1$ is the *Courant number* .





ure 1: Scheme of Leapfrog method.

“physical mode”

“computational

$$U_j^{n+1} = U_j^{n-1} - \lambda(U_{j+1}^n - U_{j-1}^n)$$

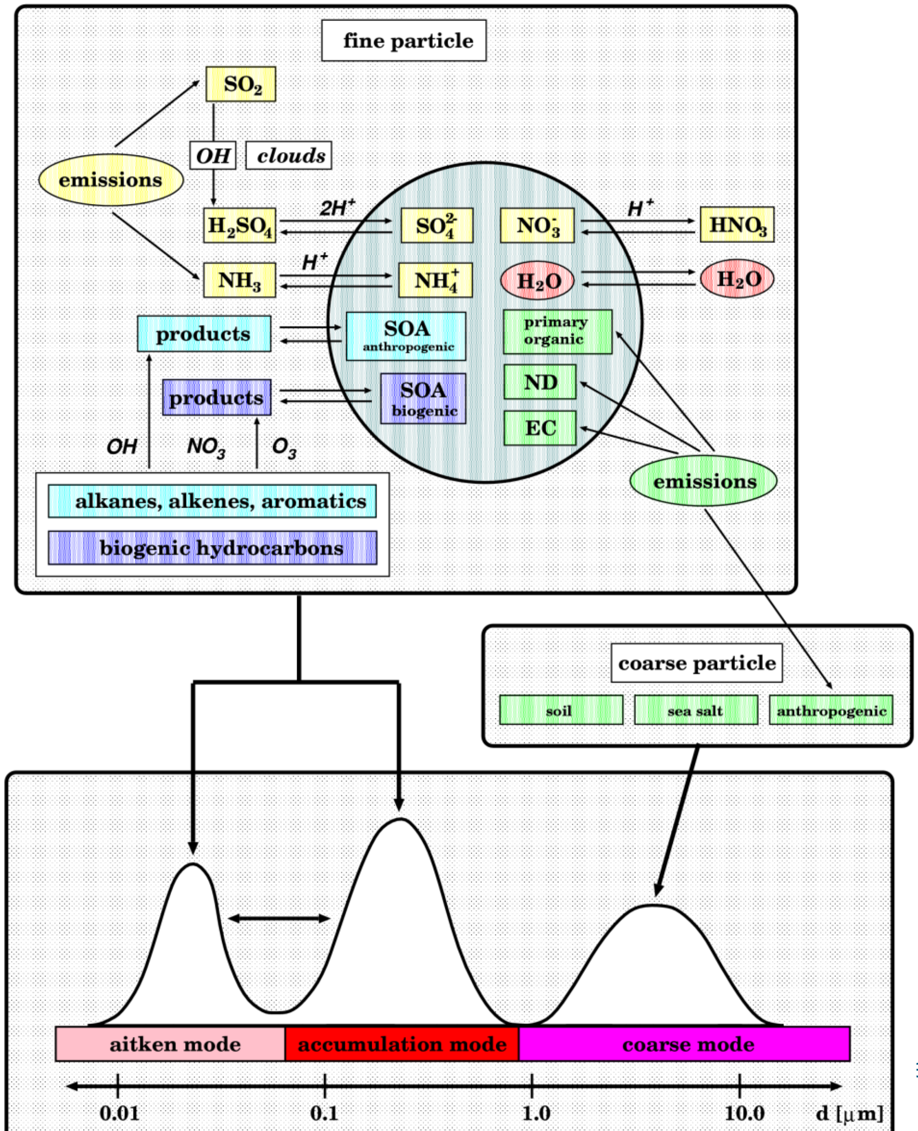
Aerosol Chemistry in MADE

Modal Aerosol Dynamics for EURAD/Europe
(Ackerman et al., 1998, Schell, 2000)

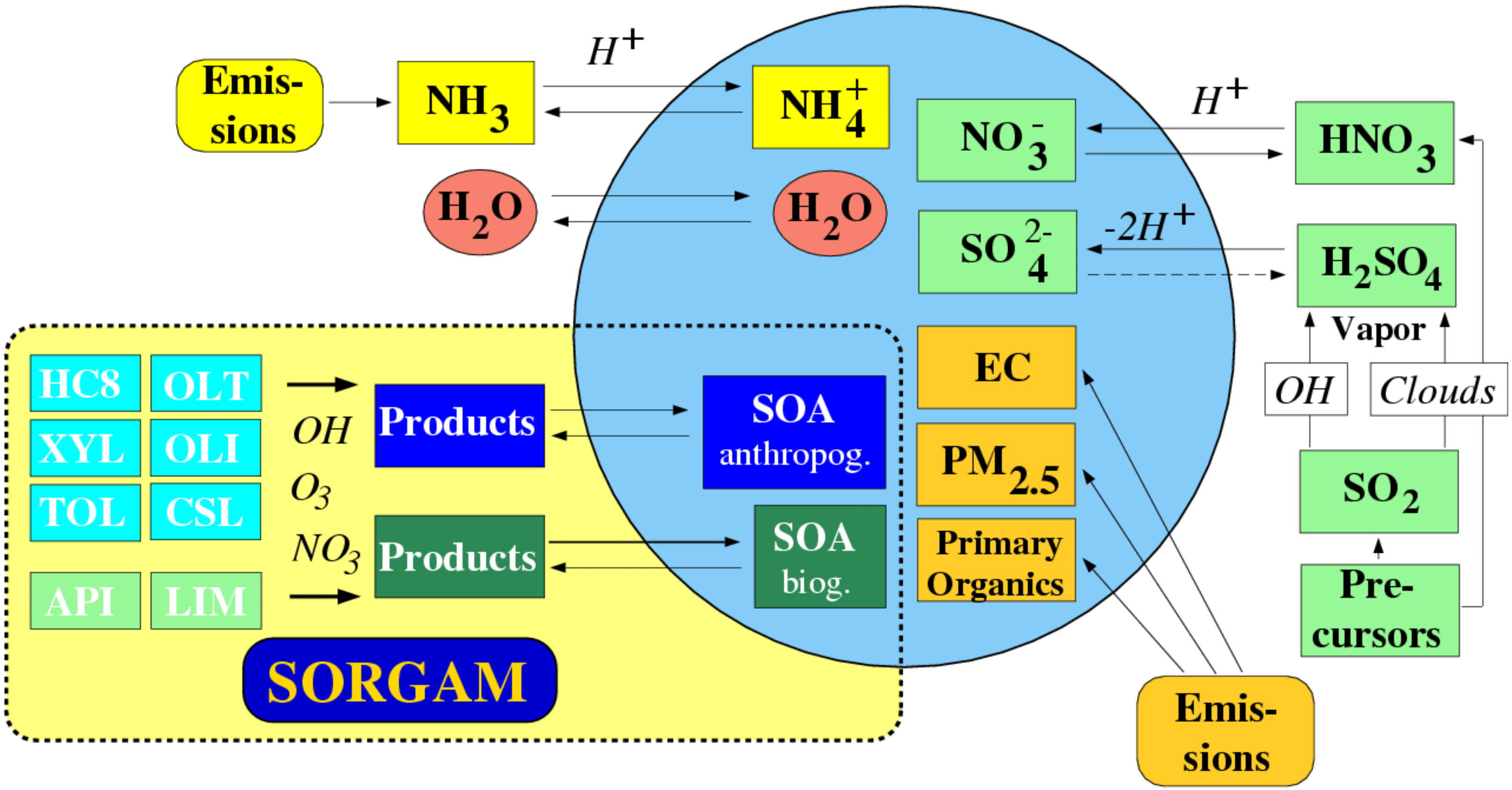
<u>Aerosol dynamics (included)</u>
Coagulation,
nucleation,
condensation-evaporation
Cloud-aerosol interaction
Diffusion, advection
Dry deposition,
sedimentation

$$dM_i^k/dt = nuk_i^k + coag_{ii}^k + coag_{ij}^k + cond_i^k + sink_i^k + emi_i^k$$

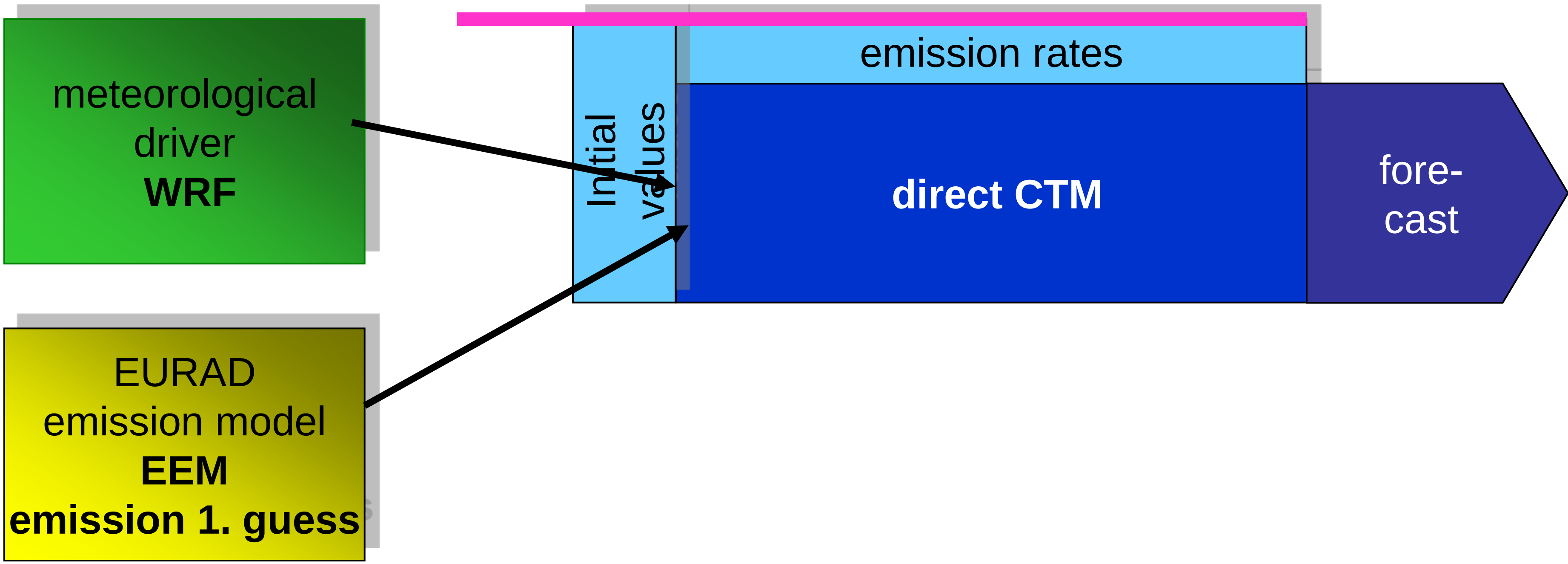
$M_i^k := k^{th}$ Moment of i^{th} Mode



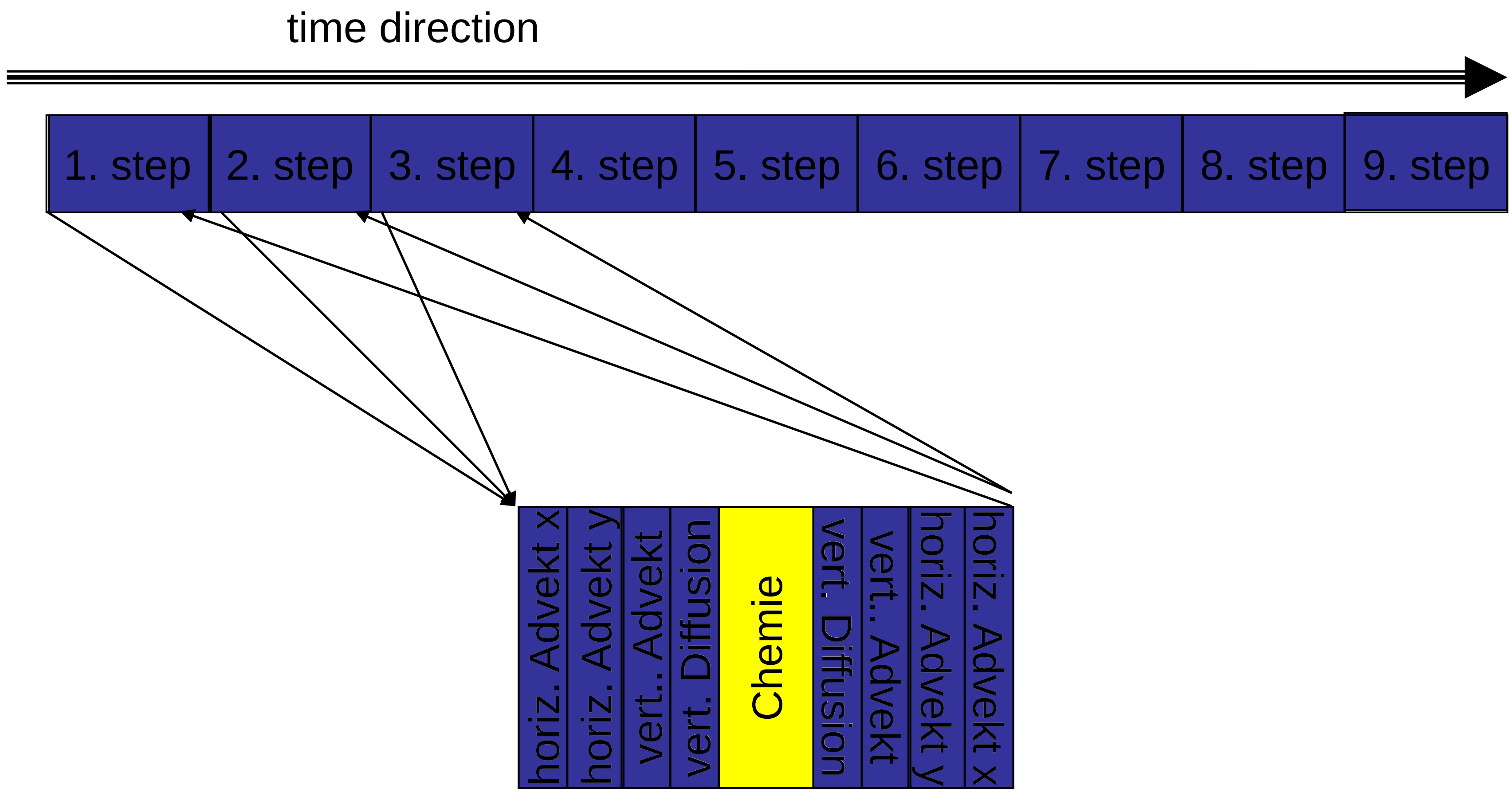
Example: chemical complexity:
The EURAD Secondary ORGanic Aerosol Model (SORGAM)



Mesoscale EURAD 4D-var data assimilation system

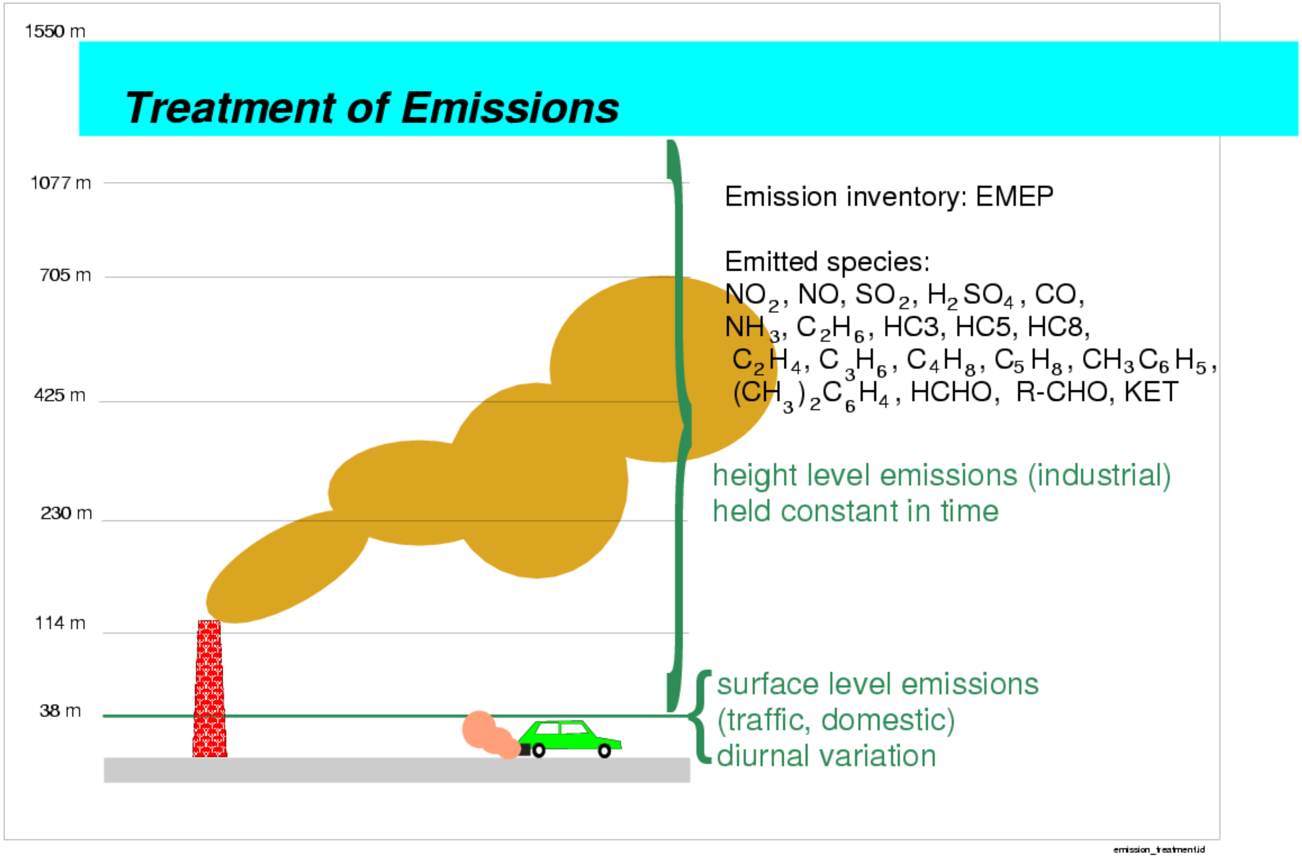


Symmetric operator split procedure



Emission pattern design (Erna)

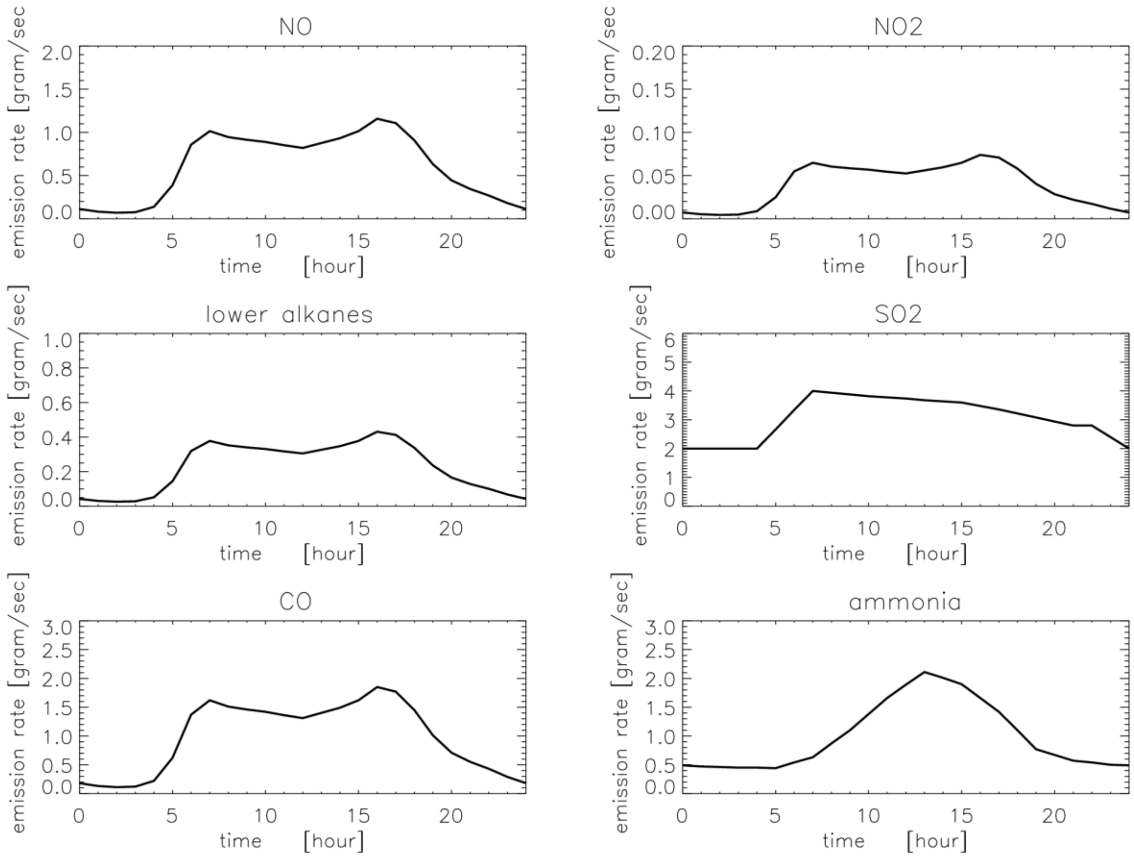
Treatment of the inverse problem for emission rate inference



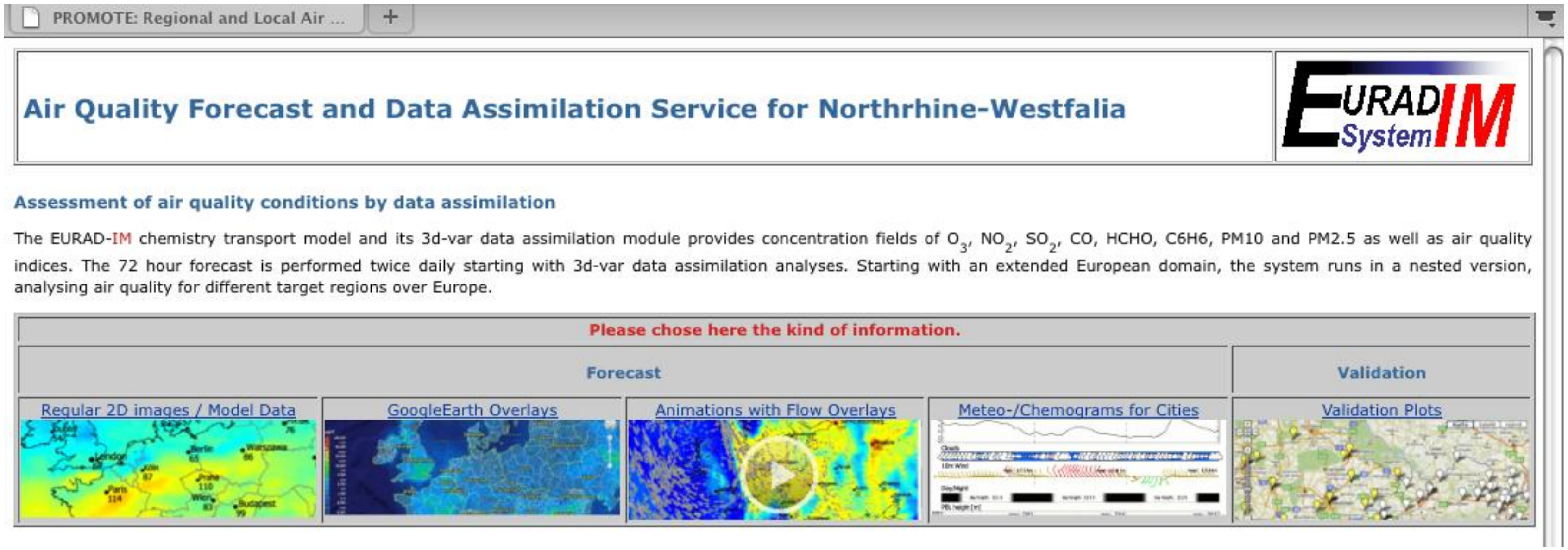
Normalised diurnal cycle of anthropogenic surface emissions *f(t)*

*emission(t)=f(t;location,species,day) * v(location,species)*

day in {working day, Saturday, Sunday} ***v optimization parameter***



Example:
Web based public user access to monitoring and forecast products for Northrine-Westfalia

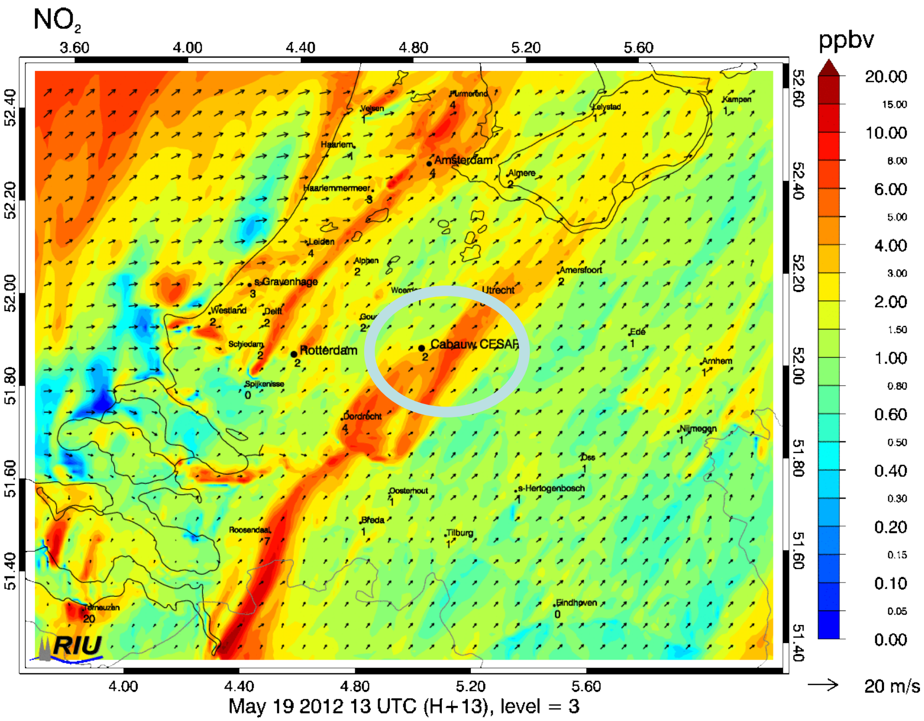


Forecast products selection panel

- 2D images (O_3, NO_2, SO_2, CO, PM_{10}, $PM_{2.5}$, air quality index)
- Google earth overlays
- Animations with flow overlay (O_3, NO_2, and PM_{10})
- Meteo-/Chemograms (50 cities for each domain)
- Validation (time-series, scatter plots)

http://db.eurad.uni-koeln.de/promote/RLAQS/riu_rlaqs.php?force=NRW

Chemical forecasts for field mission support: Example: EU FP7 PEGASOS capaign



Cabauw CESAR tower at 2012 05 19, photo by Arnoud Apituley

Zeppelin flying vertical profiles at the Cabauw tower in the observed Antwerp urban plume, as predicted by EURAD-IM (1 km resolution nest in MACC-PASODOBLE) 19.5.2012.

http://db.eurad.uni-koeln.de/ZEPTER/PEGASOS/browse_cur.php

Overview on existing models

type	scale*
global models	
regional models	$1000\text{km} \times 1000\text{km}$
mesoscale models	$100\text{km} \times 100\text{km}$
microscale models	$<< 100\text{km}$

Some regional CTMs

CMAQ (US)
http://www.cmaq-model.org/index.cfm

CAMx (US)
http://www.camx.com/

NAME (UK Met Office) Lagrangian
http://www.metoffice.gov.uk/environment/name_iii.html

WRF Chem (US)
http://ruc.noaa.gov/wrf/WG11/

CHIMERE (France)
http://www.lmd.polytechnique.fr/chimere/

Polyphemus (FRANCE)
http://cerea.enpc.fr/polyphemus/

LOTOS-EUROS (Netherlands)
http://www.lotos-euros.nl/

MOCAGE (FRANCE)
http://www.cnrm.meteo.fr/gmgec/spip.php?article87&lang=fr

EURAD-IM (Germany)
http://www.riu.uni-koeln.de (presently under modification)

What needs to be done ?

– Reducing of uncertainties of CTM

∗ emissions

∗ atmospheric transport

∗ microphysics of multiphase processes (involving the aerosol and liquid phase)

∗ chemistry of the homogeneous phases

– Coupling to climate models

∗ earth system modeling