Lecture 2
Pollution studies

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EC and ESA objectives related to operational atmospheric chemistry monitoring missions (GMES type)

- Pollutants and chemical products affecting
  - Human health
  - Growth of crops
  - condition of forests, lakes, and other small scale damageable ecosystems
  - condition of buildings

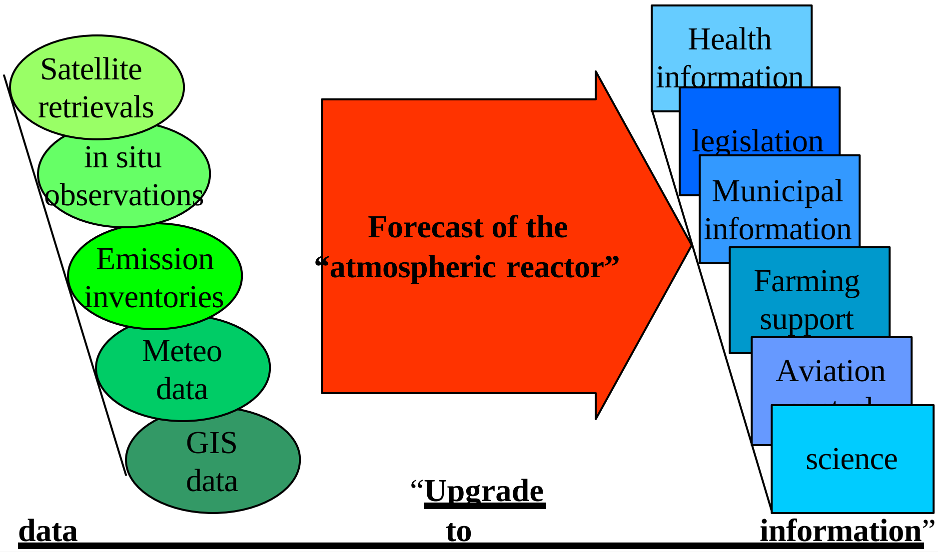
"Value added" data for operational monitoring, leads to:

- **Improved air quality forecasts**
- Impact estimates of irregular and accidental releases
- Trend estimates
- Identification of knowledge/model deficiencies

Translation into **ultimate** atmospheric chemistry simulation issues *(“what to control”)*

- Human health:
  - **PM_{10}**, $PM_{2.5}$, PM_1 *(= Particulate Matter 0->x mm)*
  - POPs (*Persistent Organic Pollutants*): **PAH**s (*Polycyclic aromatic hydrocarbons*), PCBs (*PolyClorinated Biphenyls*), HCHs (HexaCloroHexanes), **benzene, benzopyrene**, ….
  - Trace metals: **Cd**, Be, Co, Hg, Mo, **Ni**, Se, Sn, V, **As**, Cr, Cu, Mn, Zn, **Pb**
  - **Ozone**, PAN, **NO, NO_2, SO_2, CO**
  - Pollen
- Crops:
  - Ozone
- Forests, lakes, ecosystems
  - “Acid rain”
  - ozone

General objective for air pollution information processing



Satellite retrievals

in situ observations

Emission inventories

Meteo data

GIS data

Forecast of the “atmospheric reactor”

Health information

legislation

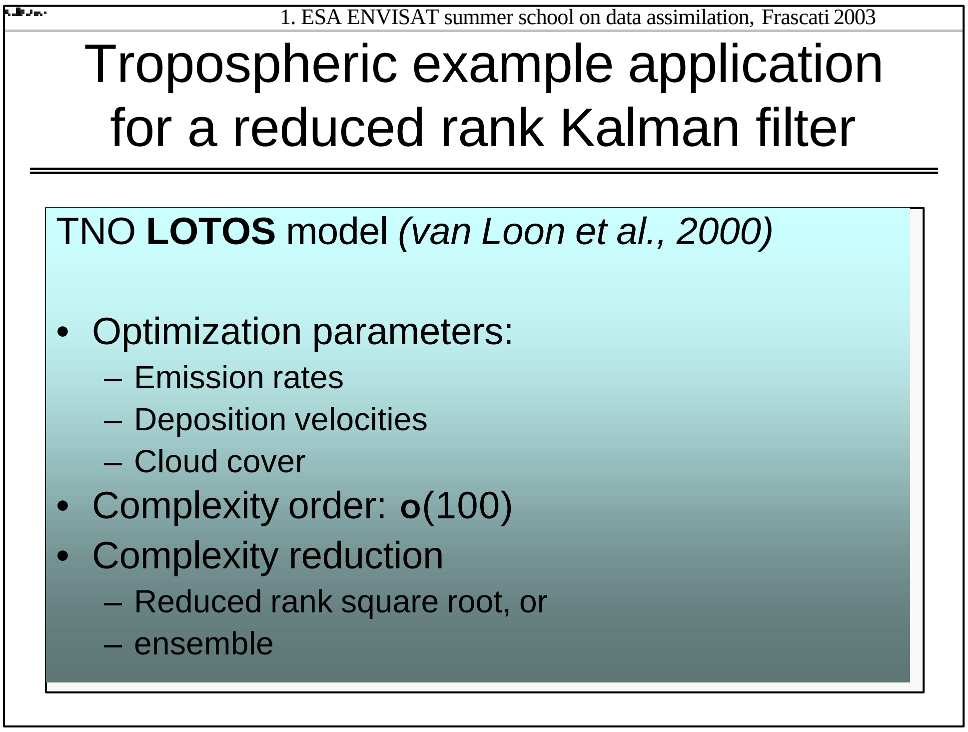
Municipal information

Farming support

Aviation

science

“**Upgrade data to information**”



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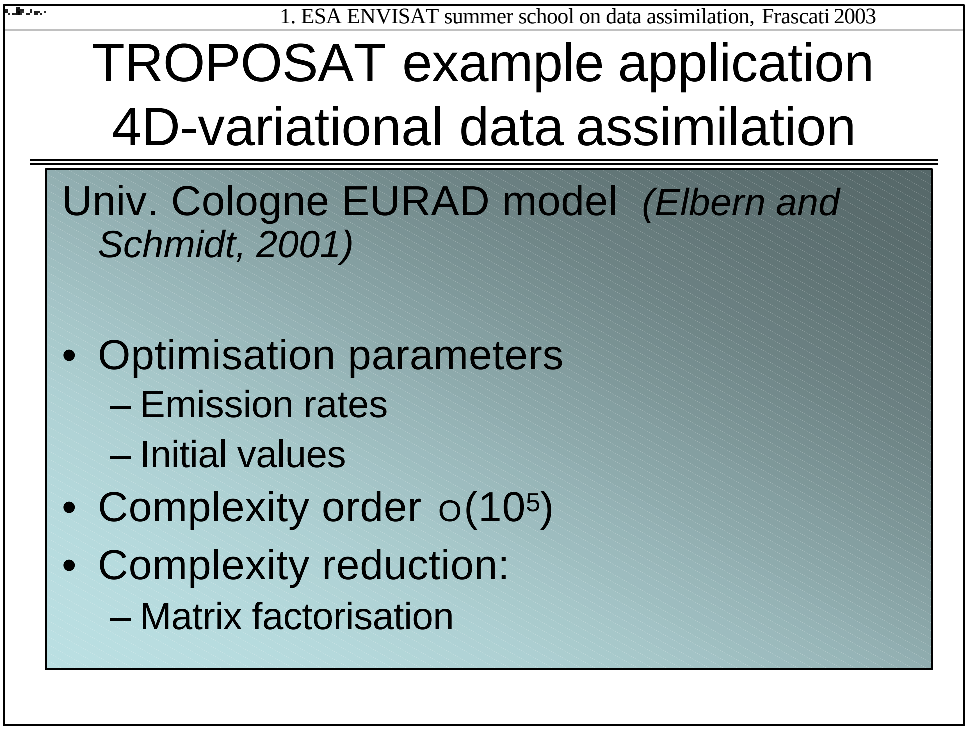
Tropospheric example application for a reduced rank Kalman filter

TNO **LOTOS** model *(van Loon et al., 2000)*

- Optimization parameters:
  - Emission rates
  - Deposition velocities
  - Cloud cover
- Complexity order: **o**(100)
- Complexity reduction
  - Reduced rank square root, or
  - ensemble



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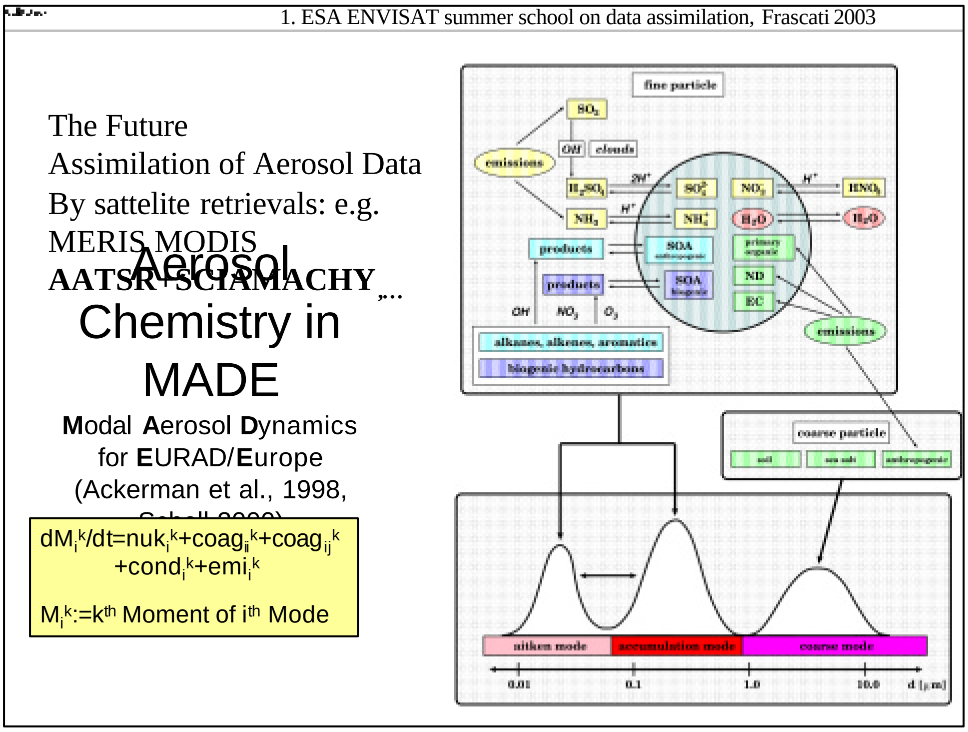
TROPOSAT example application 4D-variational data assimilation

Univ. Cologne EURAD model *(Elbern and Schmidt, 2001)*

- Optimisation parameters
  - Emission rates
  - Initial values
- Complexity order o(10^5)
- Complexity reduction:
  - Matrix factorisation



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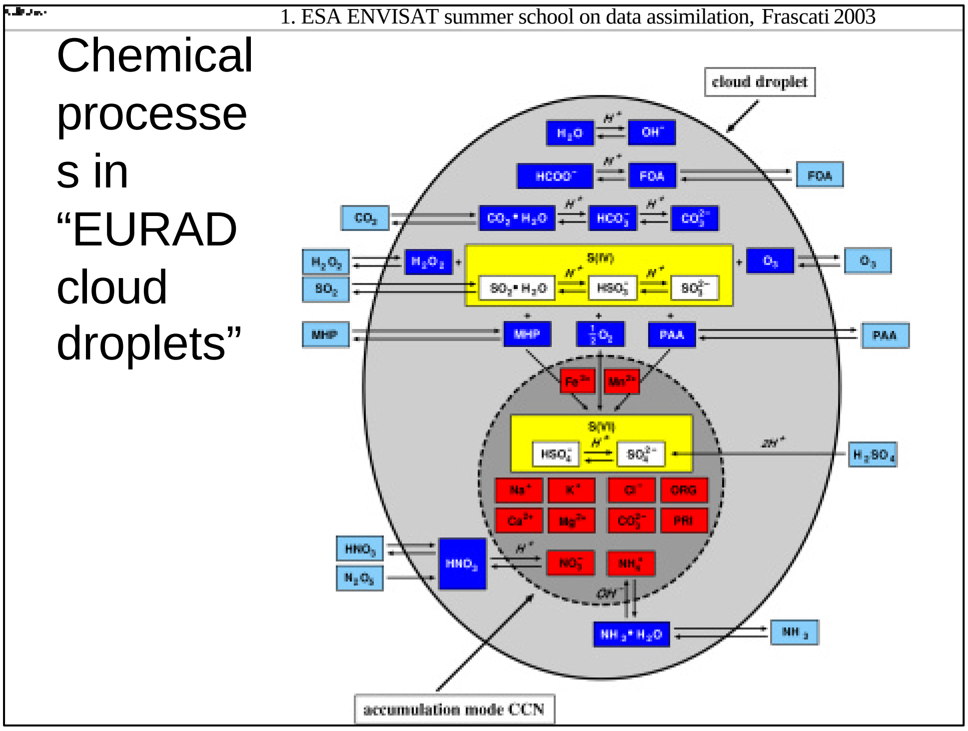
The Future
Assimilation of Aerosol Data
By sattelite retrievals: e.g.
MERIS MODIS
AATSR SCIAMACHY,...

Aerosol Chemistry in MADE

Modal **A**erosol **D**ynamics for **E**URAD/**E**urope (Ackerman et al., 1998, Schell 2000)

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

M_i^k:=kth Moment of ith Mode



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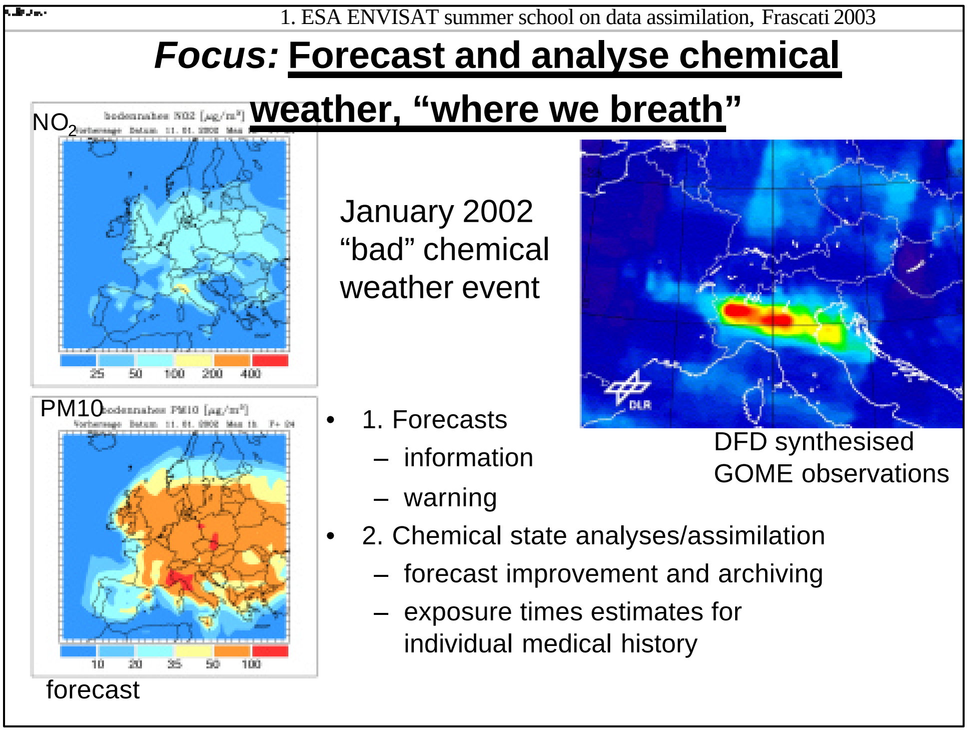
Chemical processes in “EURAD cloud droplets”



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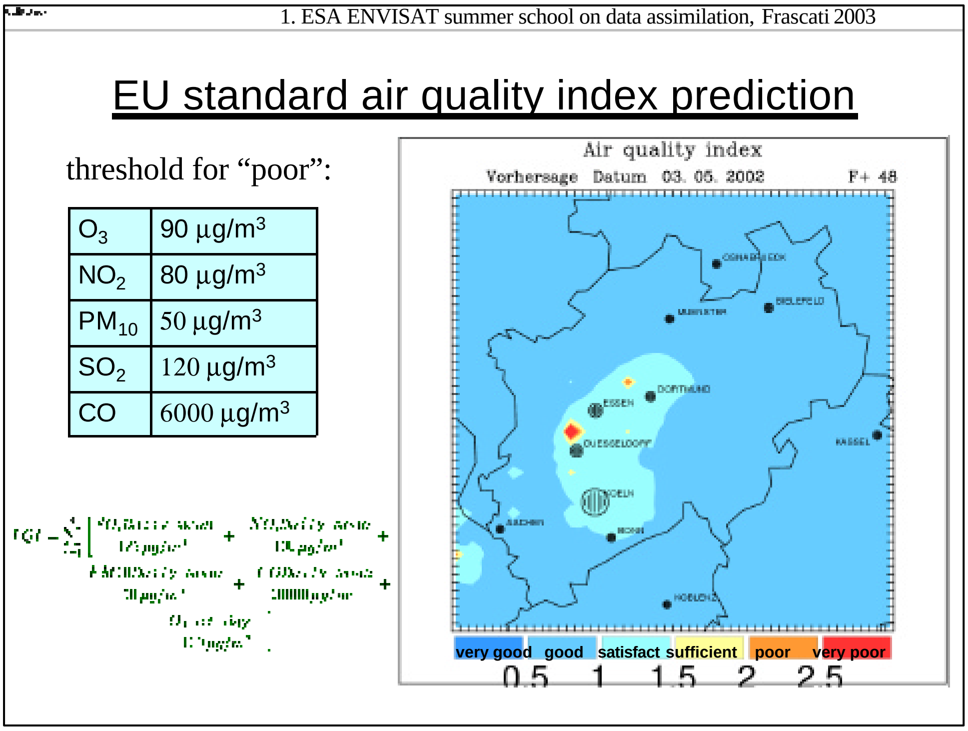
Focus: Forecast and analyse chemical weather, “where we breath”

January 2002 “bad” chemical weather event

DFD synthesised GOME observations

- 1. Forecasts
  - information
  - warning
- 2. Chemical state analyses/assimilation
  - forecast improvement and archiving
  - exposure times estimates for individual medical history

forecast



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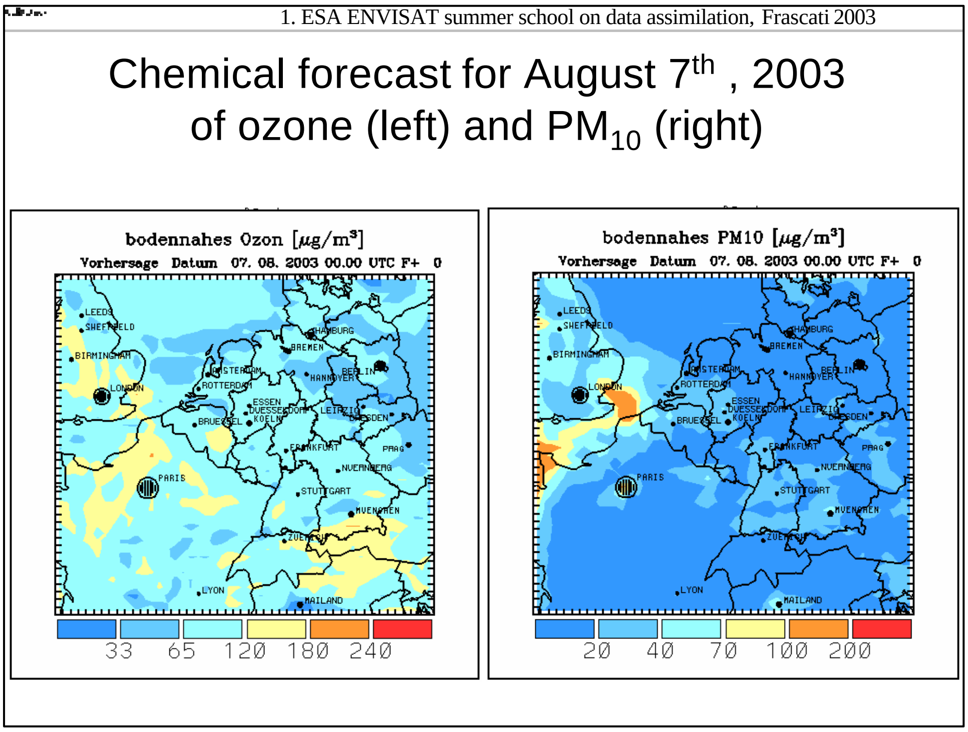
EU standard air quality index prediction

threshold for “poor”:

O_3	90 µg/m³
NO_2	80 µg/m³
PM_{10}	50 µg/m³
SO_2	120 µg/m³
CO	6000 µg/m³

[illegible]



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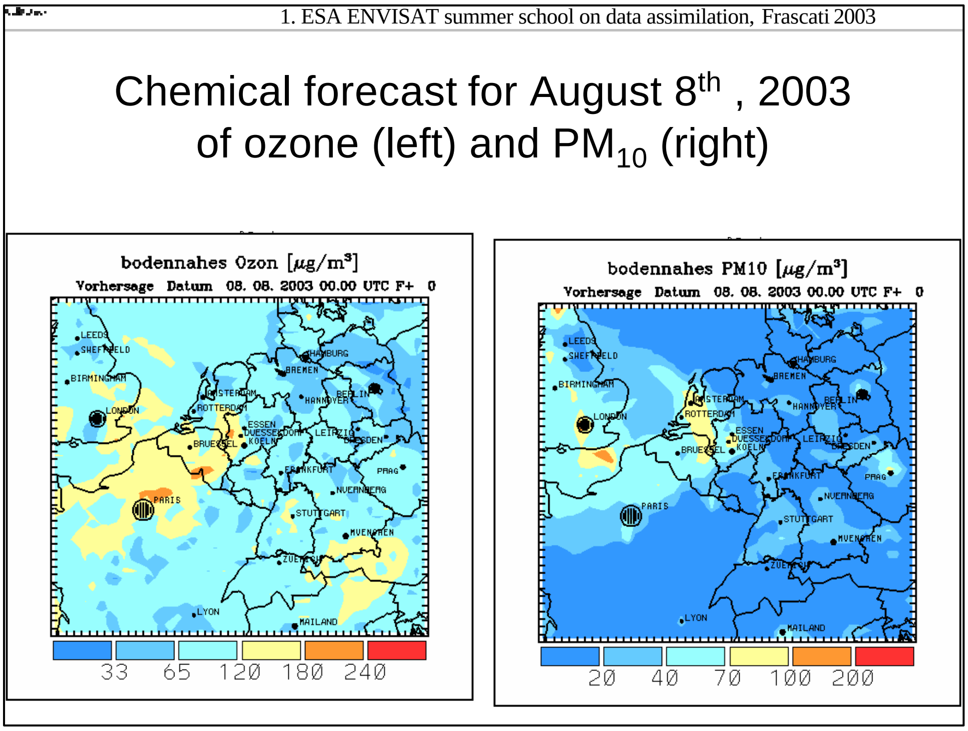
Chemical forecast for August 7th , 2003 of ozone (left) and PM_{10} (right)



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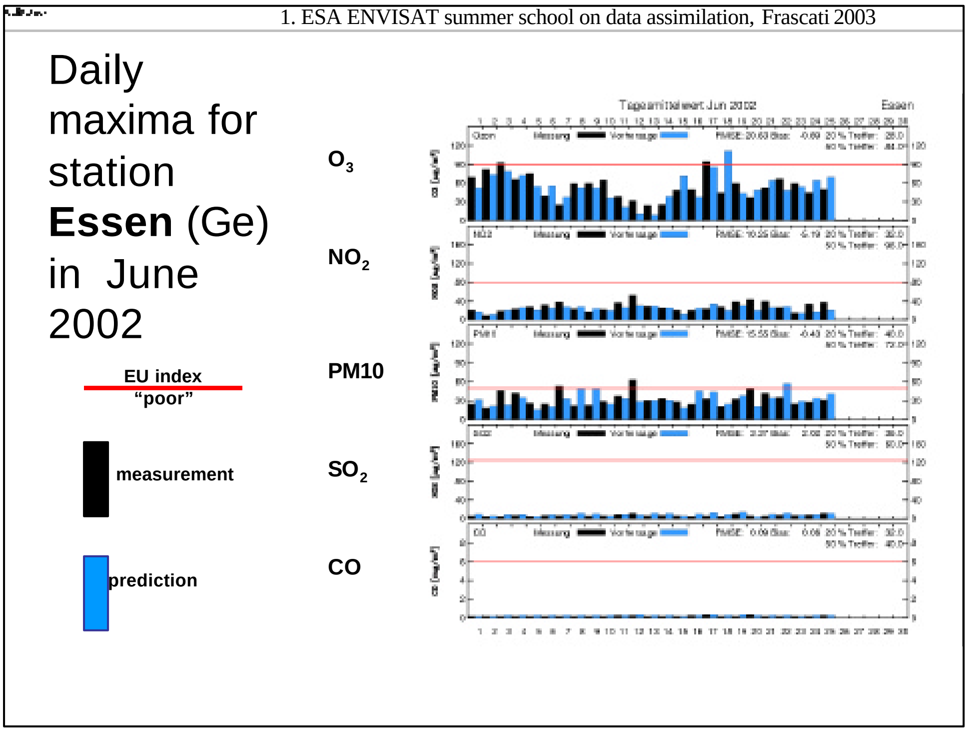
Chemical forecast for August 8th , 2003 of ozone (left) and PM_{10} (right)



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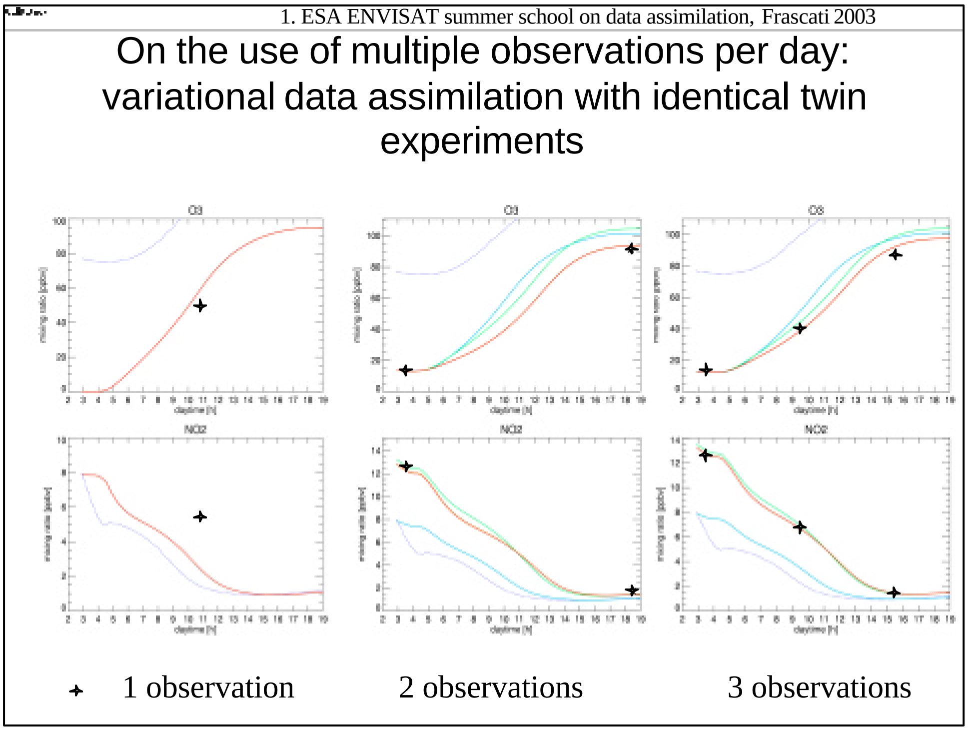
Daily maxima for station **Essen** (Ge) in June 2002

EU index "poor"

█ **measurement**

🟦 **prediction**



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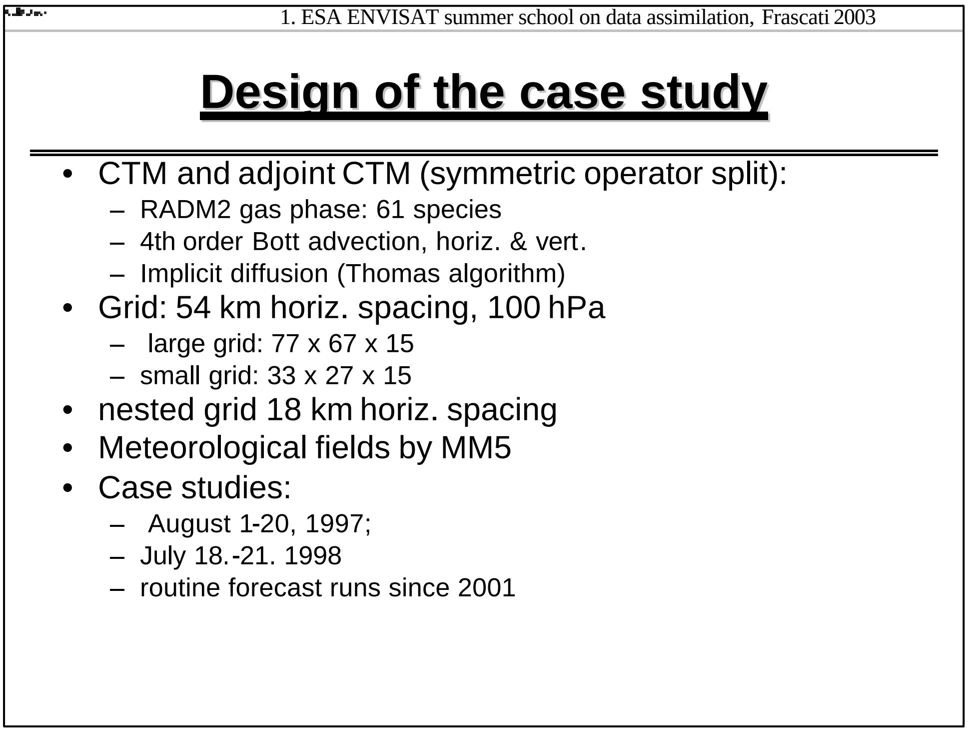
On the use of multiple observations per day: variational data assimilation with identical twin experiments

+ 1 observation

2 observations

3 observations



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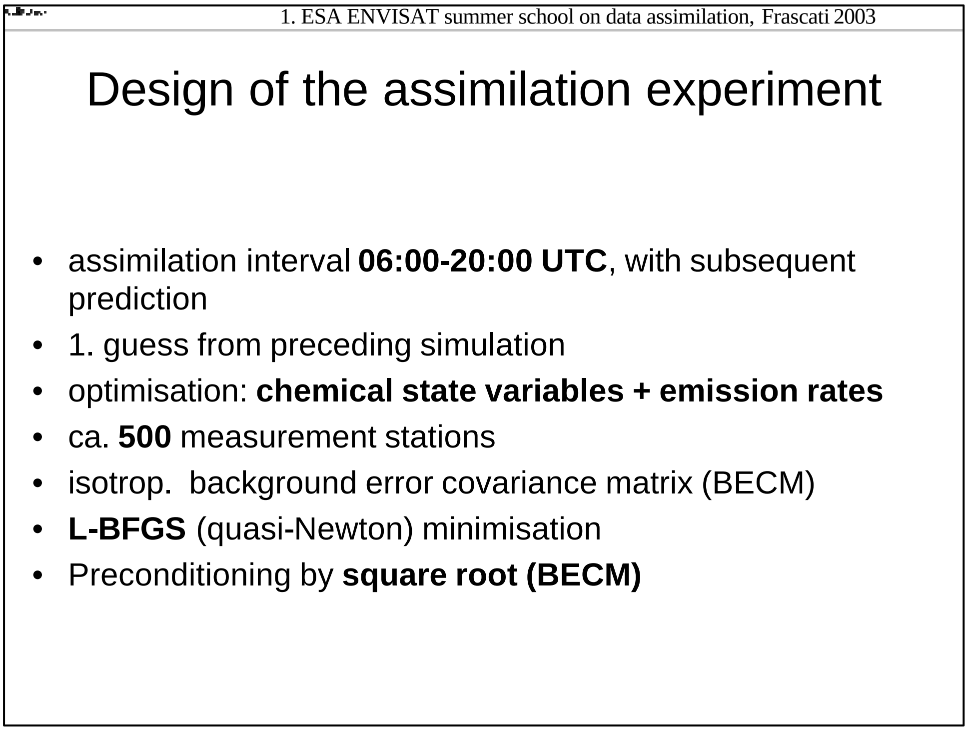
Design of the case study

- CTM and adjoint CTM (symmetric operator split):
  - RADM2 gas phase: 61 species
  - 4th order Bott advection, horiz. & vert.
  - Implicit diffusion (Thomas algorithm)
- Grid: 54 km horiz. spacing, 100 hPa
  - large grid: 77 x 67 x 15
  - small grid: 33 x 27 x 15
- nested grid 18 km horiz. spacing
- Meteorological fields by MM5
- Case studies:
  - August 1-20, 1997;
  - July 18.-21. 1998
  - routine forecast runs since 2001



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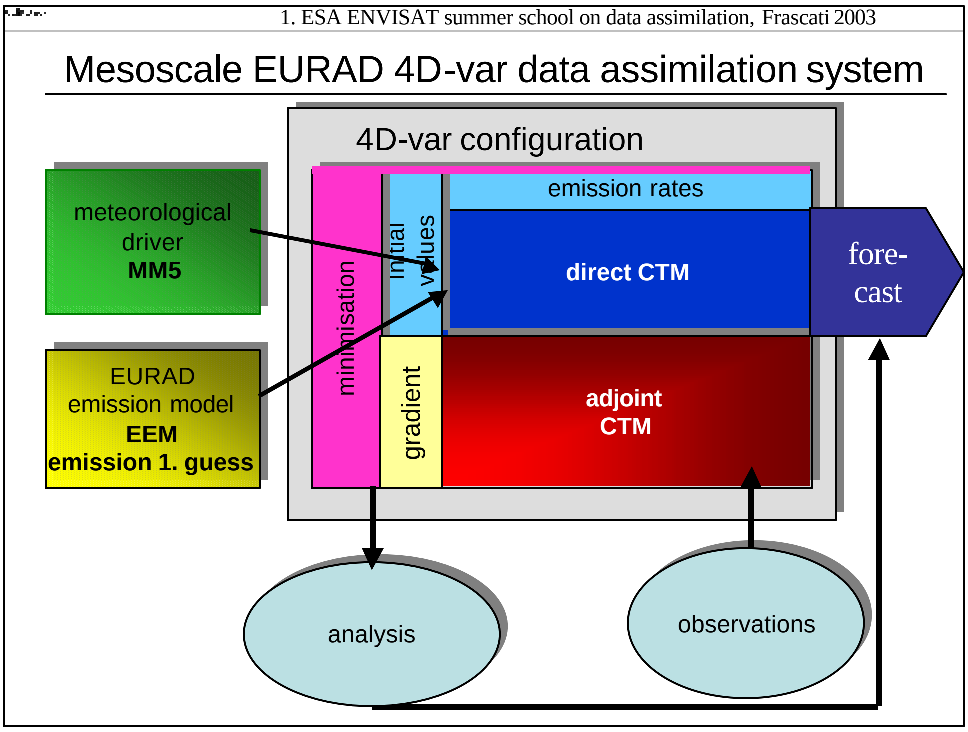
Design of the assimilation experiment

- assimilation interval **06:00-20:00 UTC**, with subsequent prediction
- 1. guess from preceding simulation
- optimisation: **chemical state variables + emission rates**
- ca. **500** measurement stations
- isotrop. background error covariance matrix (BECM)
- **L-BFGS** (quasi-Newton) minimisation
- Preconditioning by **square root (BECM)**



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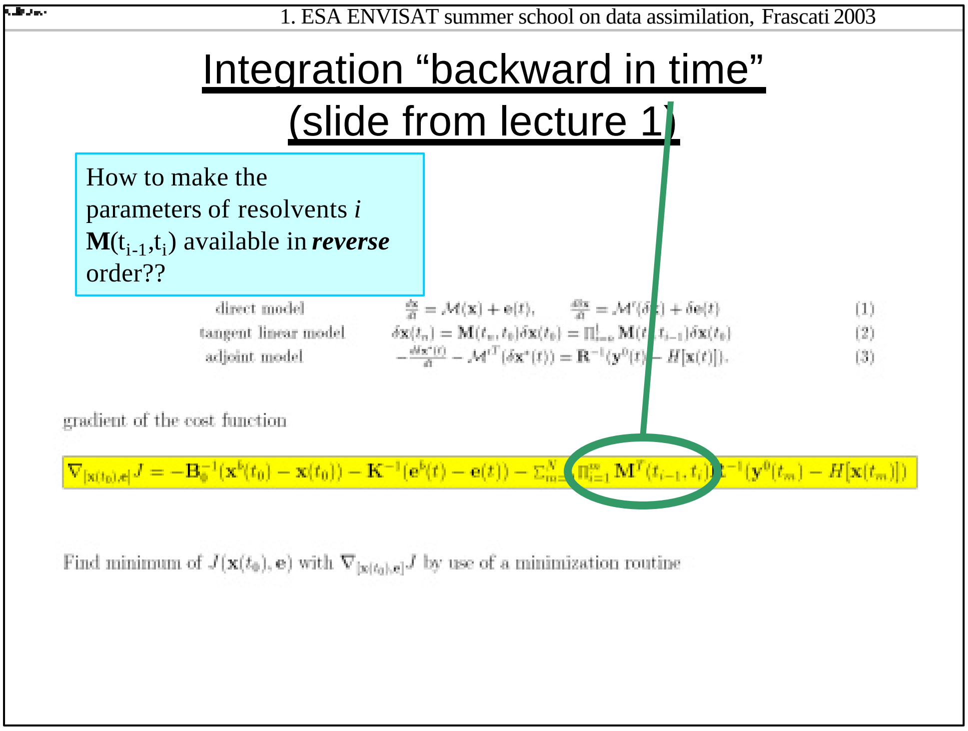
Mesoscale EURAD 4D-var data assimilation system



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Integration "backward in time" (slide from lecture 1)

How to make the parameters of resolvents *i* **M**(t_{i-1},t_i) available in ***reverse*** order??

direct model $\frac{d\mathbf{x}}{dt} = \mathcal{M}(\mathbf{x}) + \mathbf{e}(t), \quad \frac{d\delta\mathbf{x}}{dt} = \mathcal{M}'(\delta\mathbf{x}) + \delta\mathbf{e}(t)$ (1)

tangent linear model $\delta\mathbf{x}(t_n) = \mathbf{M}(t_n, t_0)\delta\mathbf{x}(t_0) = \Pi_{i=0}^{n} \mathbf{M}(t_i, t_{i-1})\delta\mathbf{x}(t_0)$ (2)

adjoint model $-\frac{d\delta\mathbf{x}^*(t)}{dt} - \mathcal{M}'^T(\delta\mathbf{x}^*(t)) = \mathbf{R}^{-1}(\mathbf{y}^0(t) - H[\mathbf{x}(t)]).$ (3)

gradient of the cost function

$$\nabla_{[\mathbf{x}(t_0),\mathbf{e}]} J = -\mathbf{B}_0^{-1}(\mathbf{x}^b(t_0) - \mathbf{x}(t_0)) - \mathbf{K}^{-1}(\mathbf{e}^b(t) - \mathbf{e}(t)) - \Sigma_{m=0}^{N} \Pi_{i=1}^{m} \mathbf{M}^T(t_{i-1}, t_i)\mathbf{R}^{-1}(\mathbf{y}^0(t_m) - H[\mathbf{x}(t_m)])$$

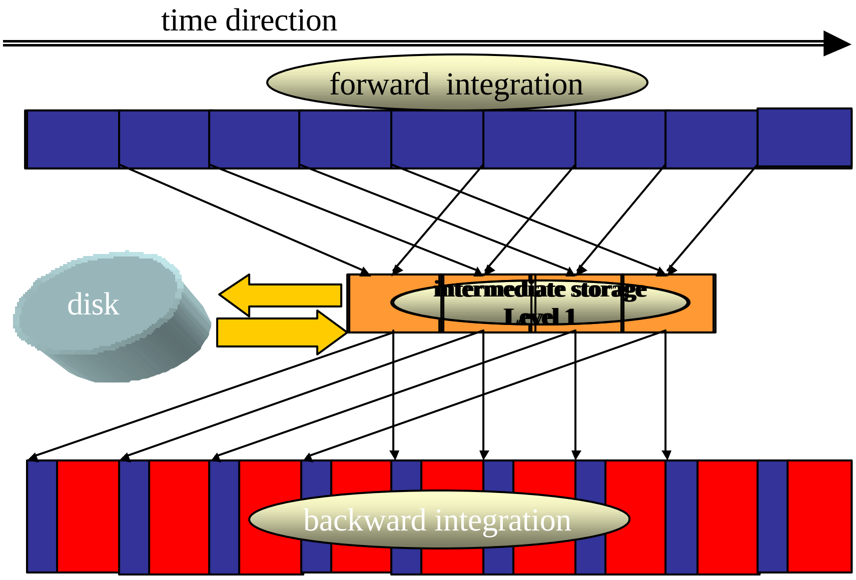
Find minimum of $J(\mathbf{x}(t_0), \mathbf{e})$ with $\nabla_{[\mathbf{x}(t_0),\mathbf{e}]} J$ by use of a minimization routine

Computational complexity estimate of the variational algorithm

Nx*Ny*Nz	spatial dimensions $O(10^4\text{-}10^5)$
N_c	# constituents O(100)
N_T	# time steps of assimilation window O(10-100)
N_o	# operators O(10)
const	intermediate results $O(10^4)$

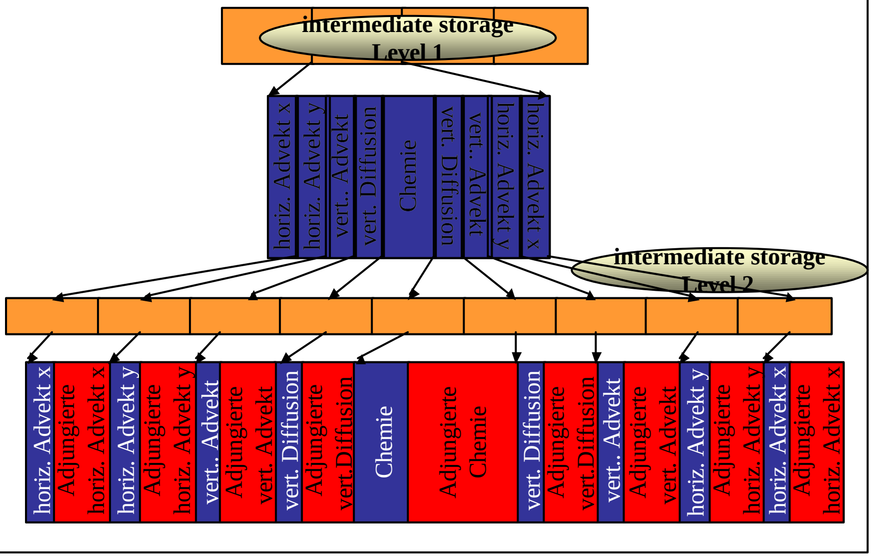
Storage strategy	# forward runs/iteration	storage	Complexity $[T_{forw}]$
total storage	1	$const*N_x*N_y*N_z*N_c*N_T*N_o$ $O(10^{12}\text{-}10^{13})$	3
operatorwise 1 level	2	$N_x*N_y*N_z*N_c*N_T*N_o$ $O(10^8\text{-}10^9)$	4
dynamic stepwise 2 levels	3	$N_x*N_y*N_z*N_c*N_T$ $O(10^7\text{-}10^8)$	5

2 level forward and backward integration scheme

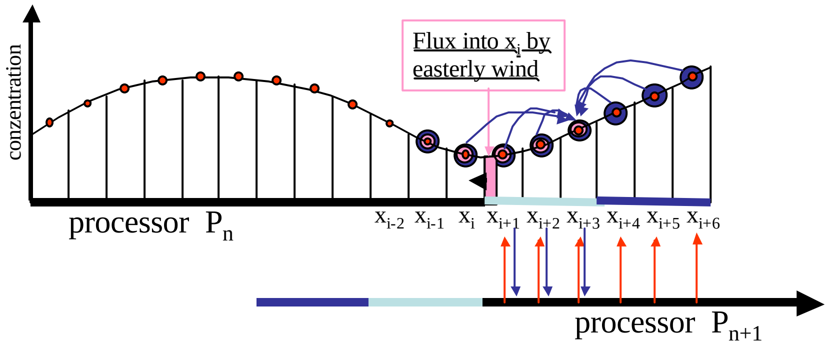


storage sequence: level 2, operator split

(each time step)



Data exchange for parallel adjoint transport



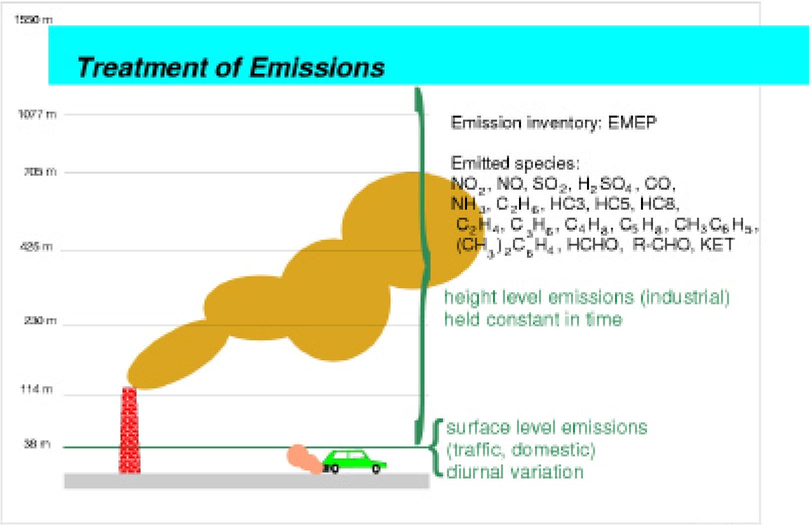
○ Grid points contributing to the calculation of the easterly flux into x_i (4th order pol.)

● Grid points contributing to the calculation of the adjoint of grid point at x_{i+3}

Computational resources requested

- with a 14 h assimilation interval about 18 iterations requested
- results in 12 CPU-hours with 121 processors of a T3E

Treatment of the inverse problem to infer emission rates



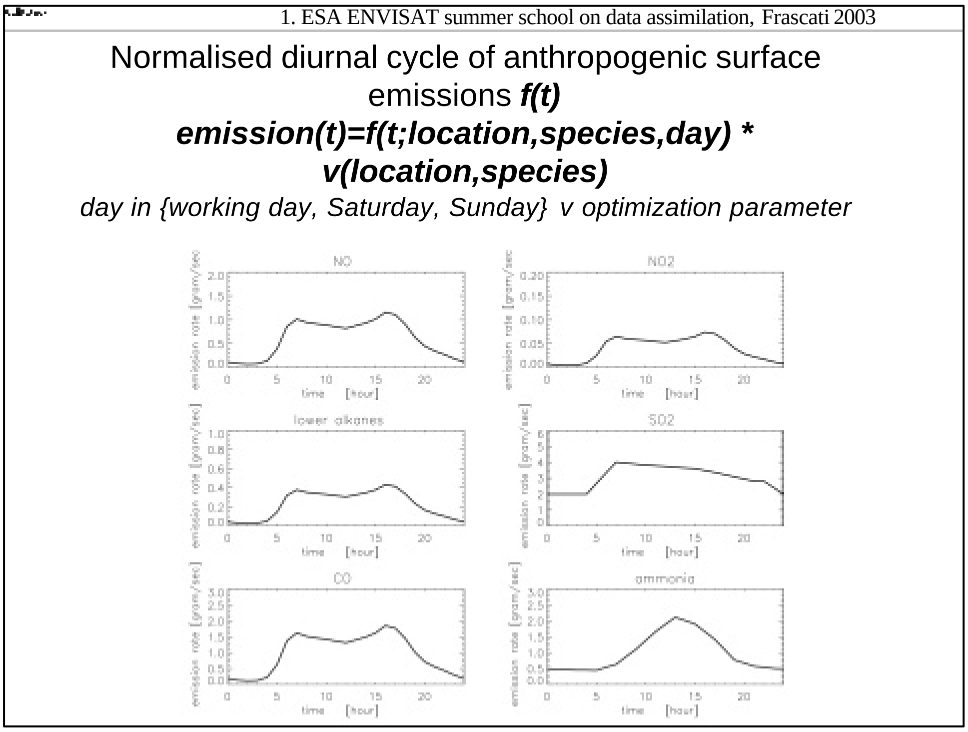
Treatment of Emissions

Emission inventory: EMEP

Emitted species:
NO_2, NO, SO_2, H_2SO_4, CO,
NH_3, C_2H_6, HC3, HC5, HC8,
C_2H_4, C_3H_6, C_4H_8, C_5H_8, $CH_3C_6H_5$,
$(CH_3)_2C_6H_4$, HCHO, R-CHO, KET

height level emissions (industrial)
held constant in time

surface level emissions
(traffic, domestic)
diurnal variation



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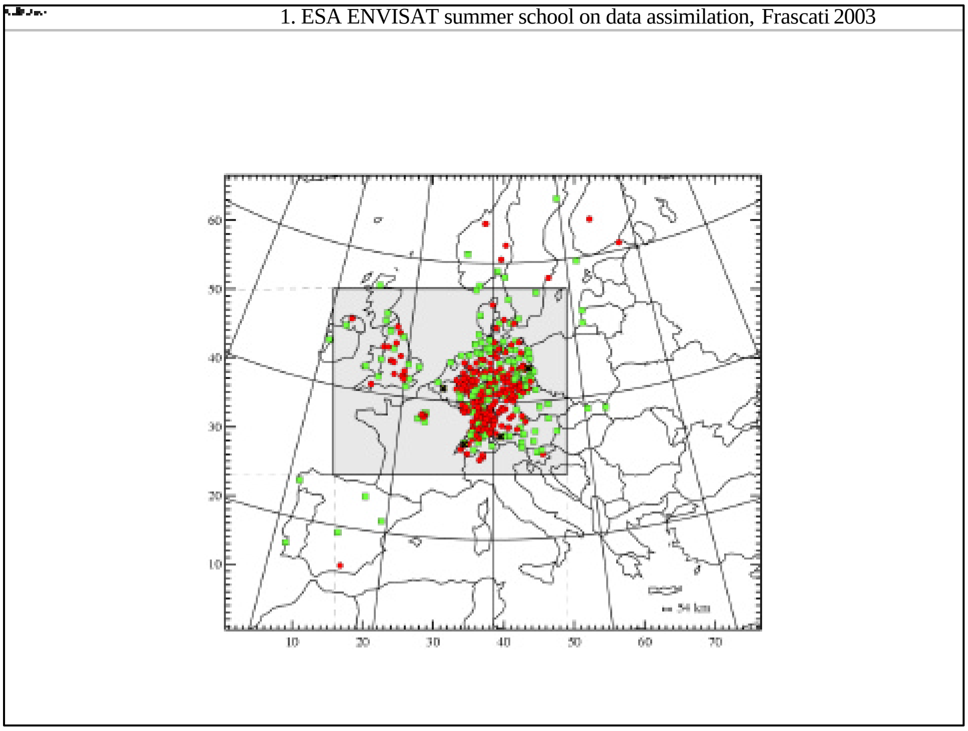
Normalised diurnal cycle of anthropogenic surface emissions ***f(t)***

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

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



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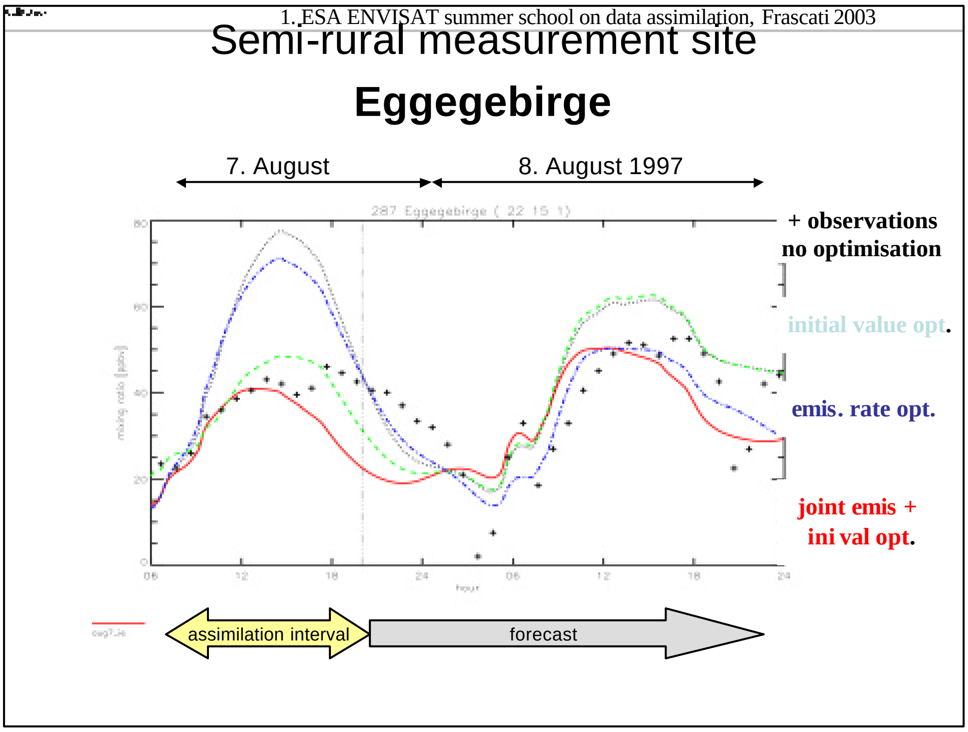

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Semi-rural measurement site

Eggegebirge

7. August

8. August 1997

+ observations

no optimisation

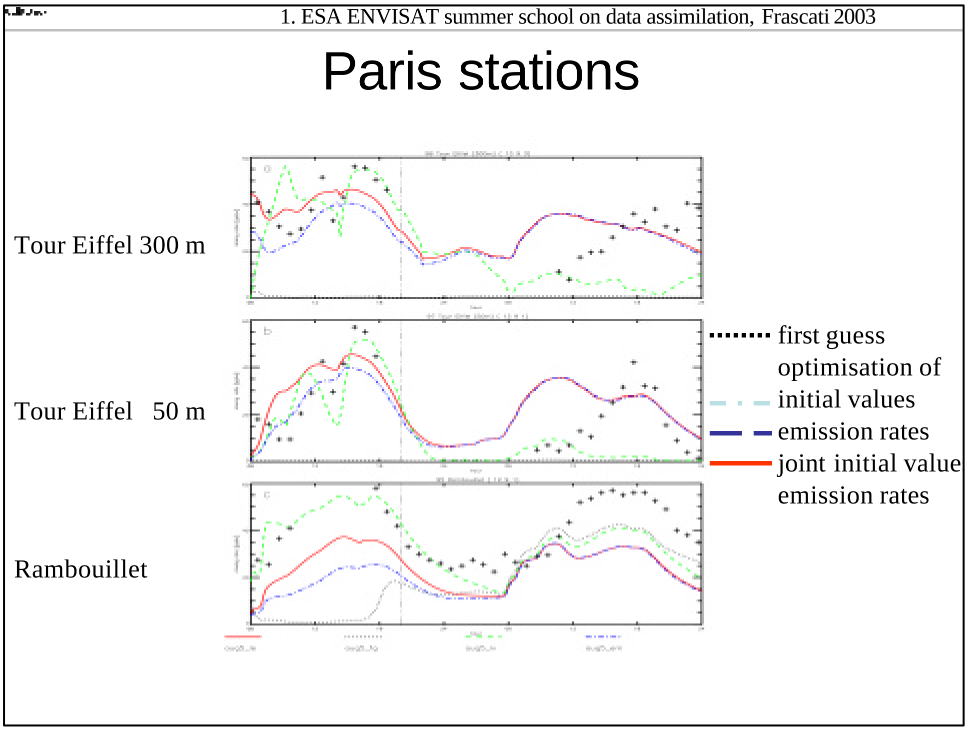
initial value opt.

emis. rate opt.

joint emis + ini val opt.

assimilation interval

forecast



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Paris stations

Tour Eiffel 300 m

Tour Eiffel 50 m

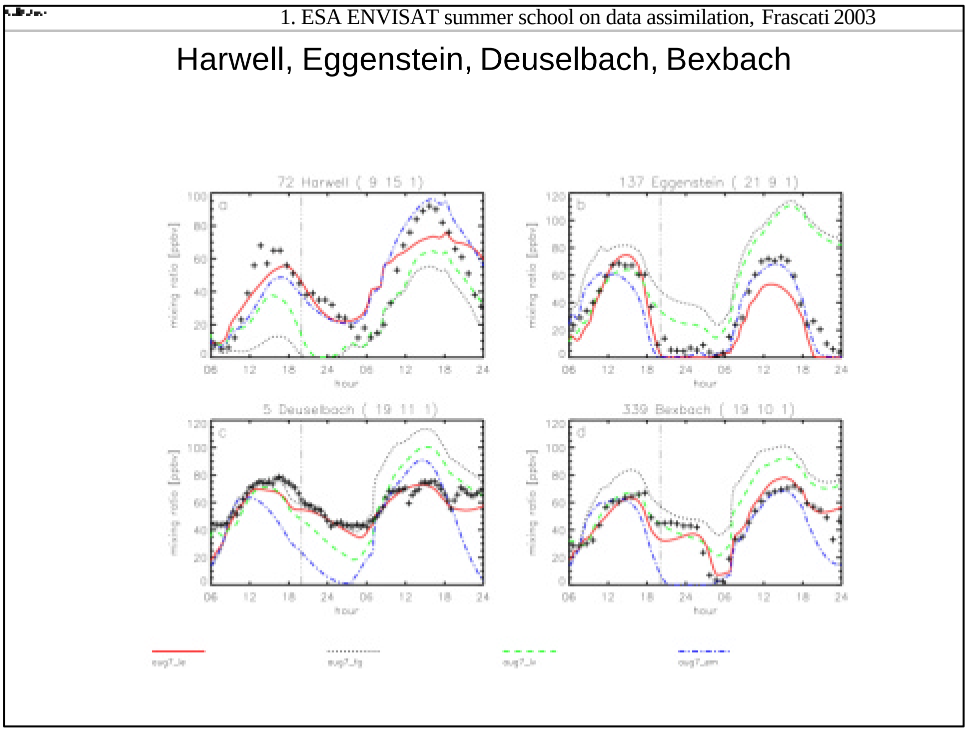
Rambouillet

first guess

optimisation of initial values

emission rates

joint initial value emission rates



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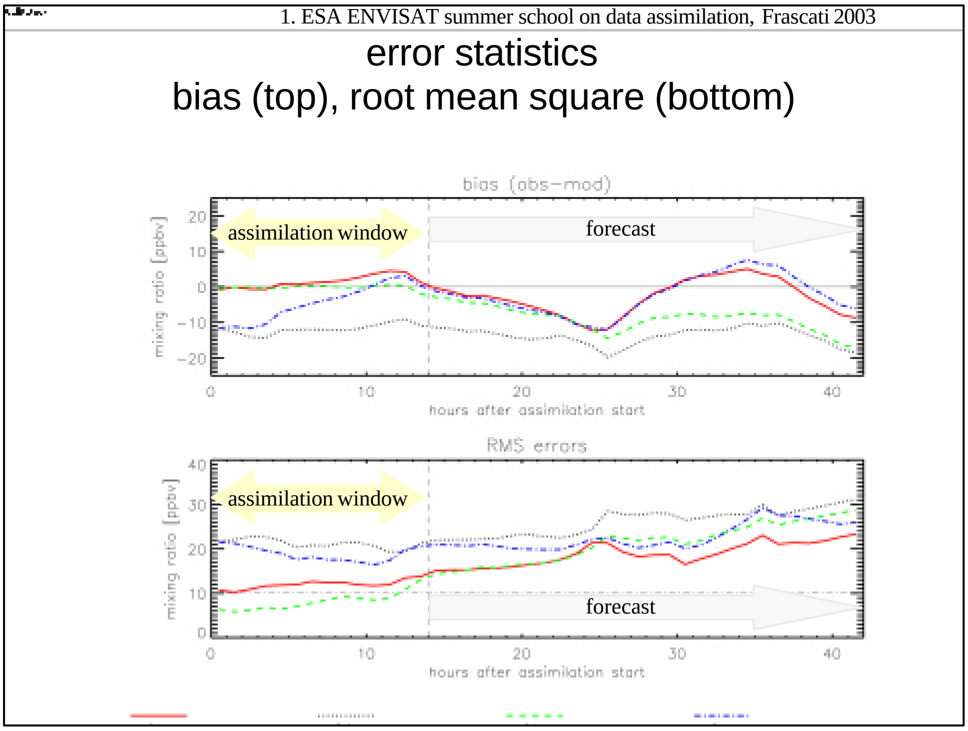
Harwell, Eggenstein, Deuselbach, Bexbach



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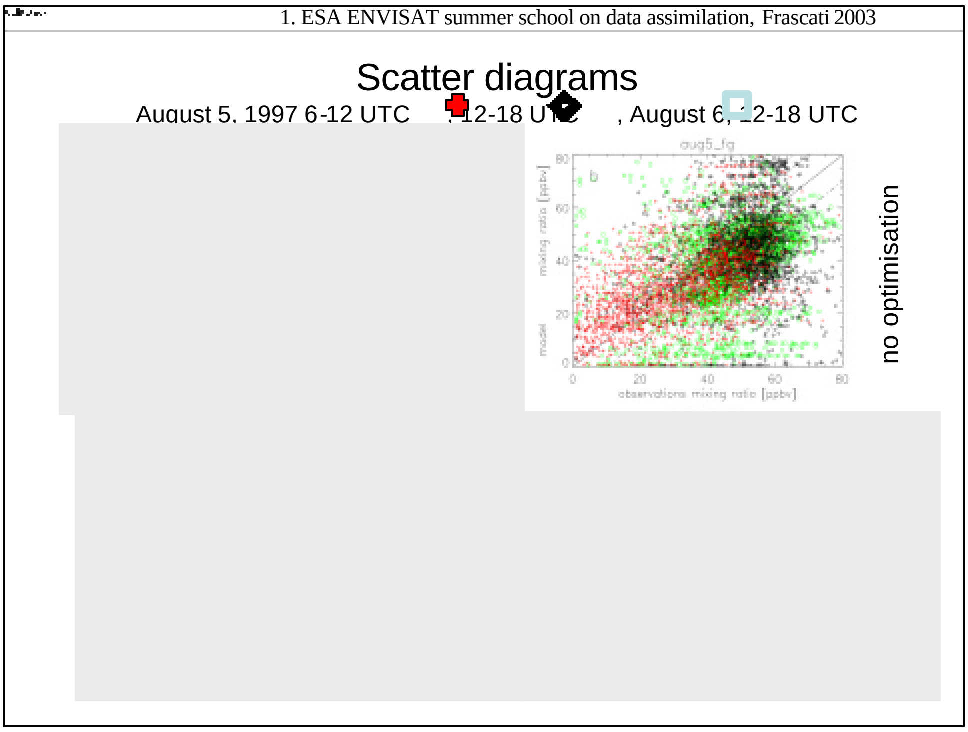
error statistics
bias (top), root mean square (bottom)



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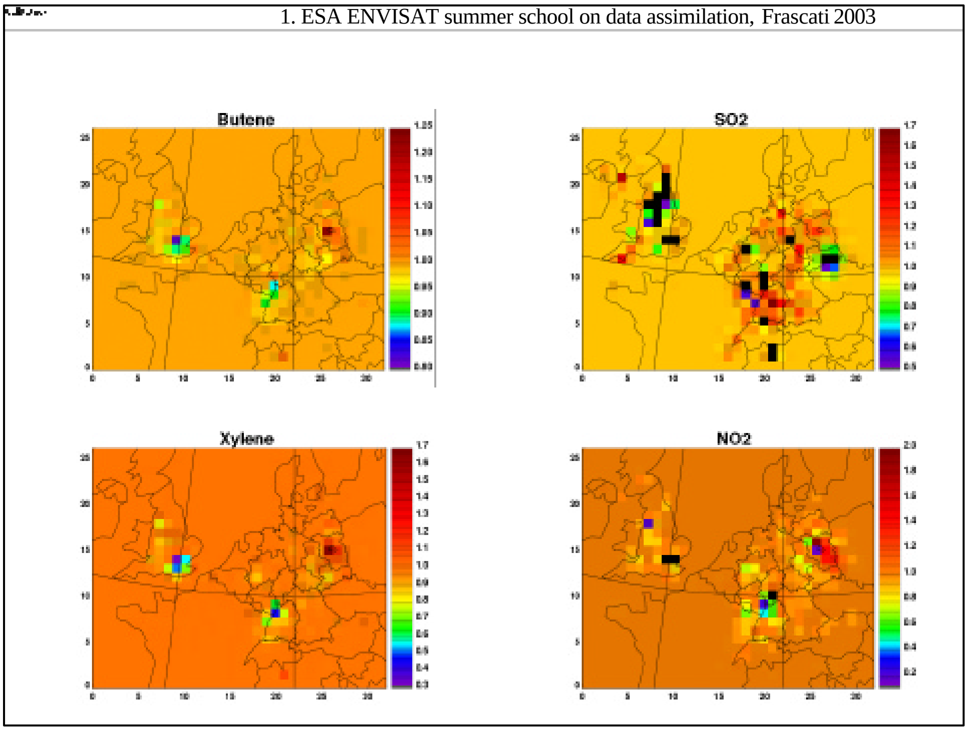
Scatter diagrams

August 5, 1997 6-12 UTC ✚, 12-18 UTC ◆, August 6, 12-18 UTC □

no optimisation



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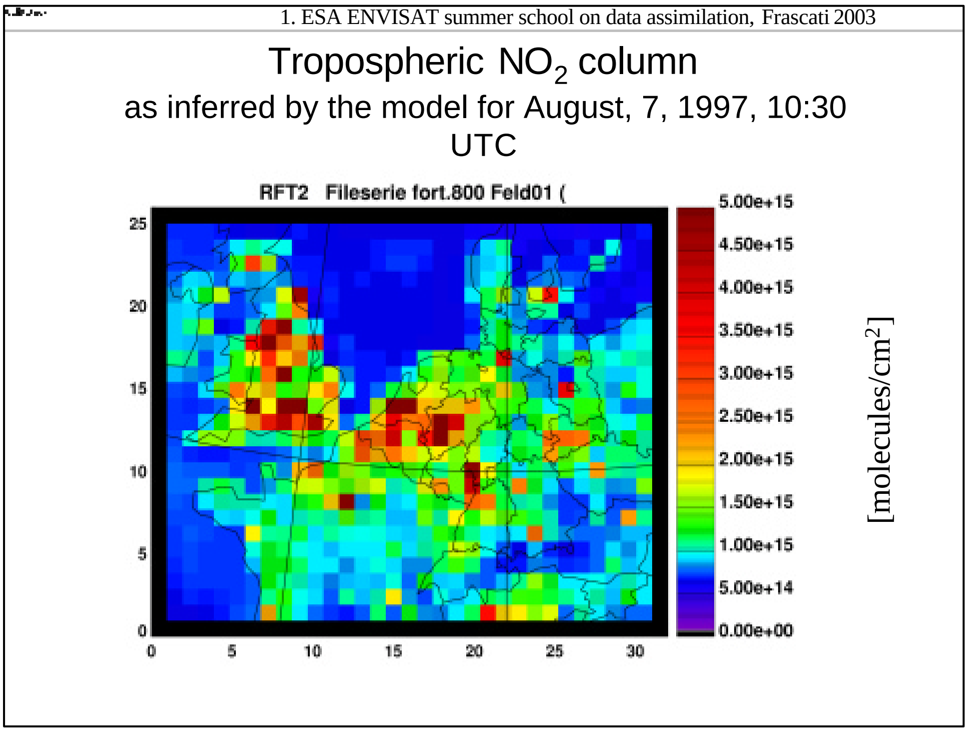

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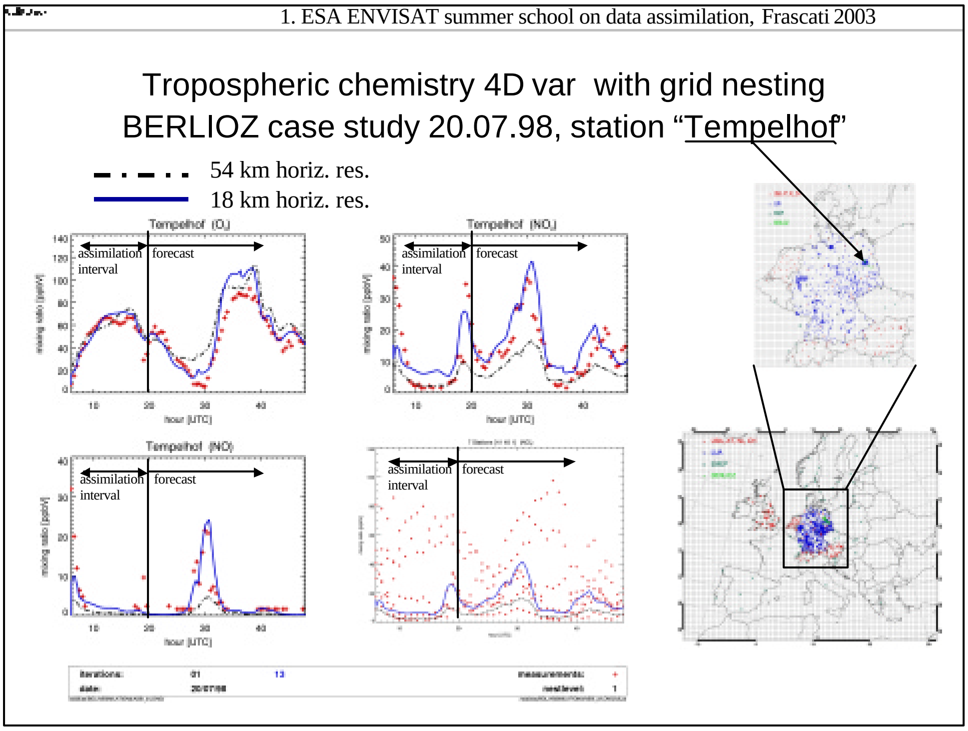
Tropospheric NO_2 column

as inferred by the model for August, 7, 1997, 10:30 UTC



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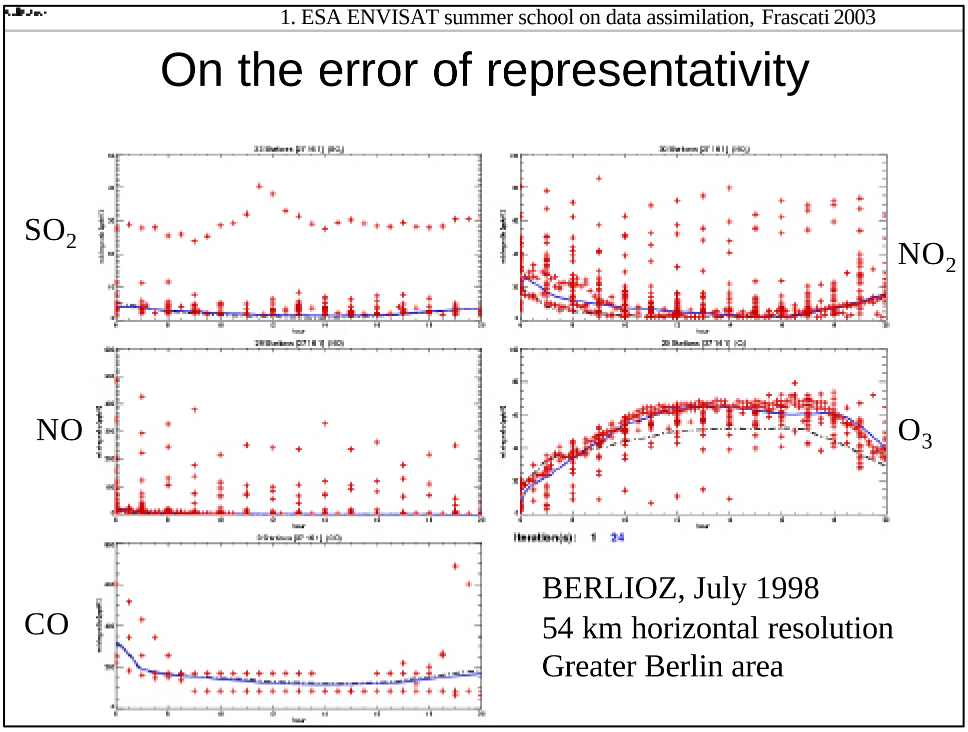
Tropospheric chemistry 4D var with grid nesting

BERLIOZ case study 20.07.98, station "Tempelhof"



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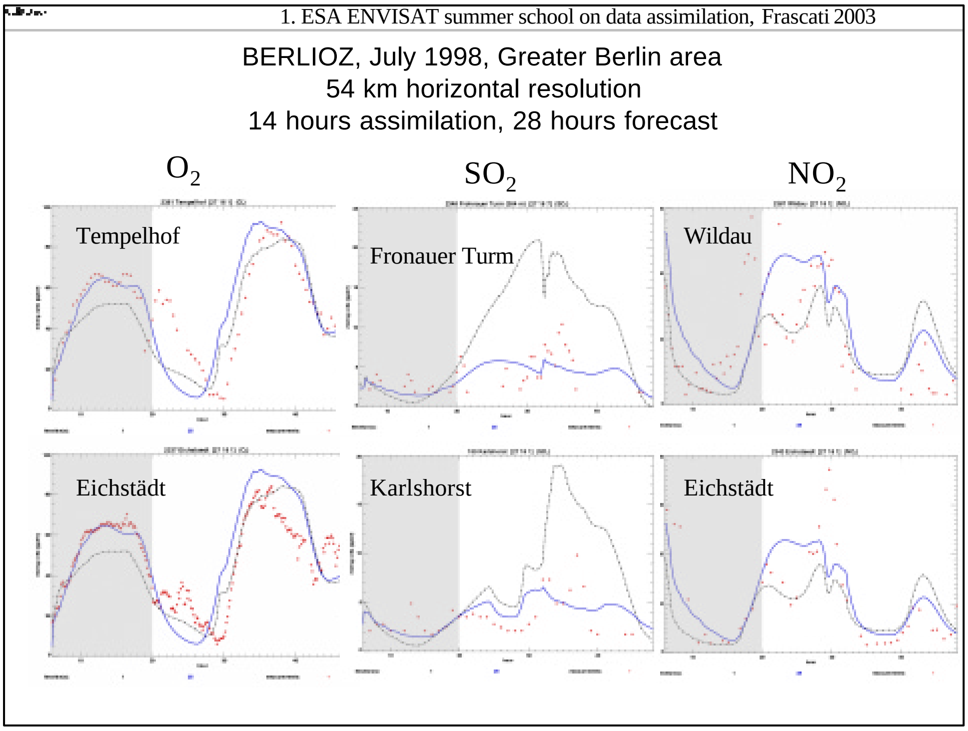
On the error of representativity

BERLIOZ, July 1998
54 km horizontal resolution
Greater Berlin area



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BERLIOZ, July 1998, Greater Berlin area
54 km horizontal resolution
14 hours assimilation, 28 hours forecast

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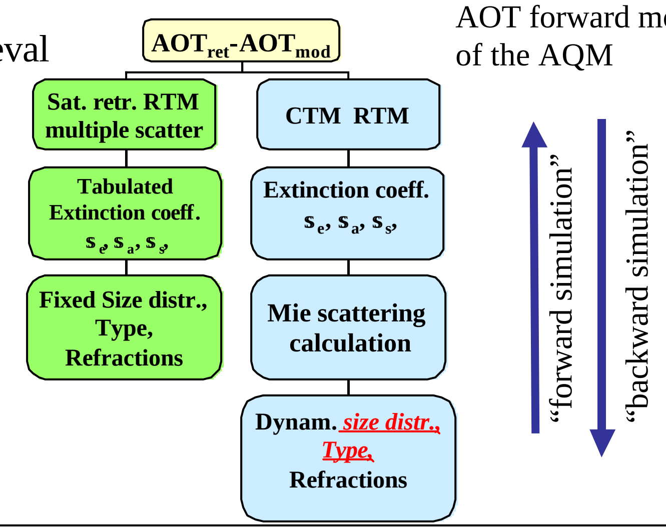
Comparing aerosol retrievals with model results for assimilation (obs-model discrepancy)

Satellite retrieval

AOT forward model of the AQM



AOT_{ret}-AOT_{mod}

Sat. retr. RTM multiple scatter

Tabulated Extinction coeff. s_e, s_a, s_s,

Fixed Size distr., Type, Refractions

CTM RTM

Extinction coeff. s_e, s_a, s_s,

Mie scattering calculation

Dynam. ***size distr., Type,*** **Refractions**

“forward simulation”

“backward simulation”

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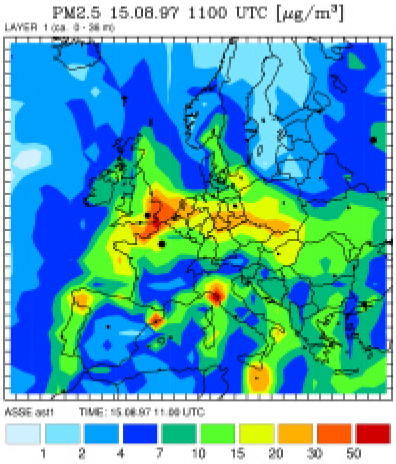
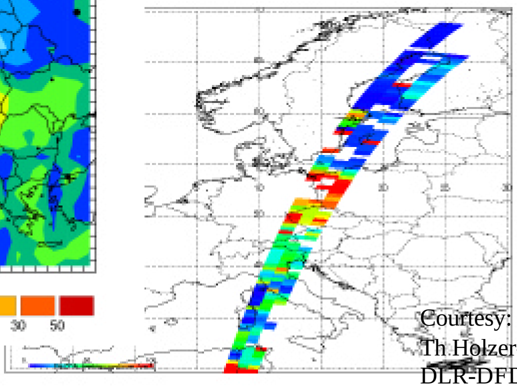
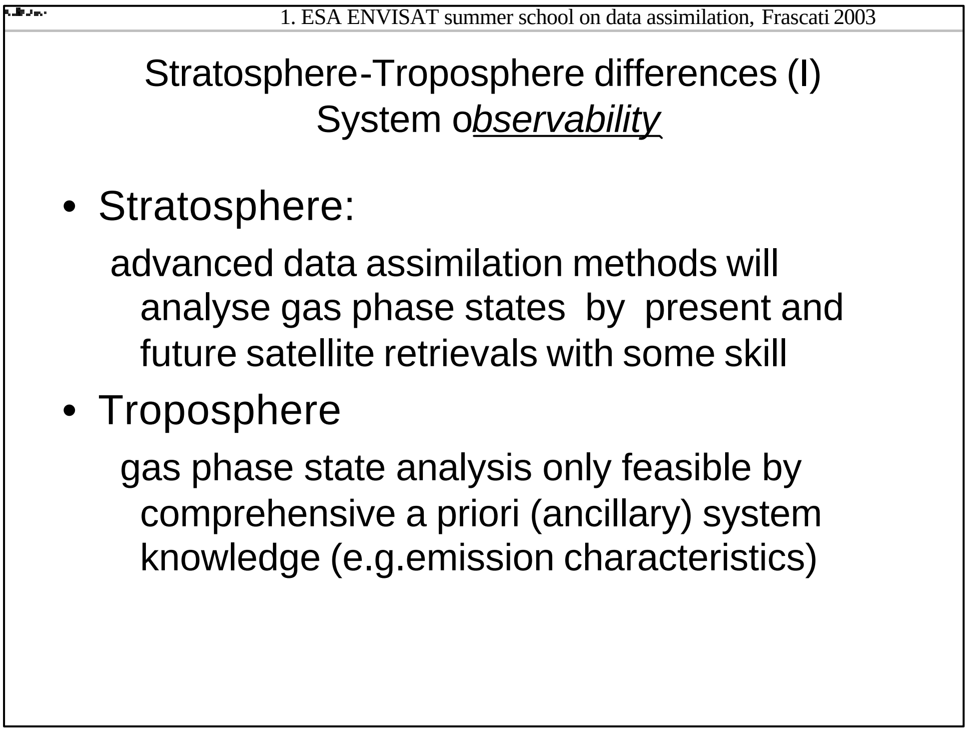
PM2.5 [µg/m³]

Note: colour code scaling different!





Courtesy: Th.Holzer-Popp, DLR-DFD



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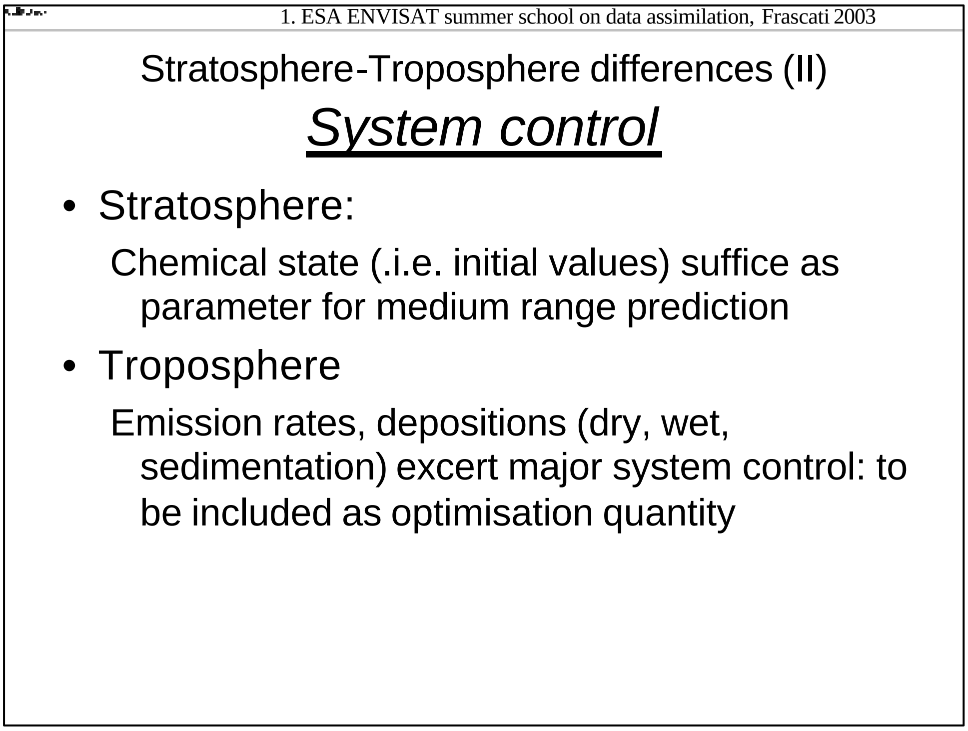
Stratosphere-Troposphere differences (I)
System o*bservability*

- Stratosphere:

  advanced data assimilation methods will analyse gas phase states by present and future satellite retrievals with some skill
- Troposphere

  gas phase state analysis only feasible by comprehensive a priori (ancillary) system knowledge (e.g.emission characteristics)



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Stratosphere-Troposphere differences (II)
System control

- Stratosphere:

  Chemical state (.i.e. initial values) suffice as parameter for medium range prediction
- Troposphere

  Emission rates, depositions (dry, wet, sedimentation) excert major system control: to be included as optimisation quantity

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Stratosphere-Troposphere differences (III)

Observation representativity

- Stratosphere:

  Representativity error of observations can be expected to be **roughly Gaussian** except at polar vortex and terminator edges (some species)

- Troposphere

  "land use" controlled, **frequently not Gaussian**

  => Finer resolution to avoid non-Gaussian methods

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PROs and CONs for variational data assimilation in tropospheric chemisty

Pro: Largely consistent combination of models and observations and other information

Improvement of forecasts and chemical state analyses

Parameter optimisation along estimated incertainties

Contra: Method is compute and development intense

Error covariances not easy to estimate: inhomogeneity and cross-parameter covariances

For significantly improved forecasts/analyses we need in the future : (aqueous chem./aerosol focus)

- Numerical complexity reduction methods
  (simulate only what really matters)
- Assimilation of observed cloud and boundary layer state
  (in routine practice: Doppler radar data both space borne and land)

Acknowledgments

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