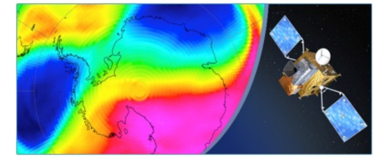
2nd ESA Advanced Training Course on Atmospheric remote Sensing
27-31 October 2014, Forschungszentrum Jülich, Germany



Regional Data assimilation

in atmospheric chemistry and air quality

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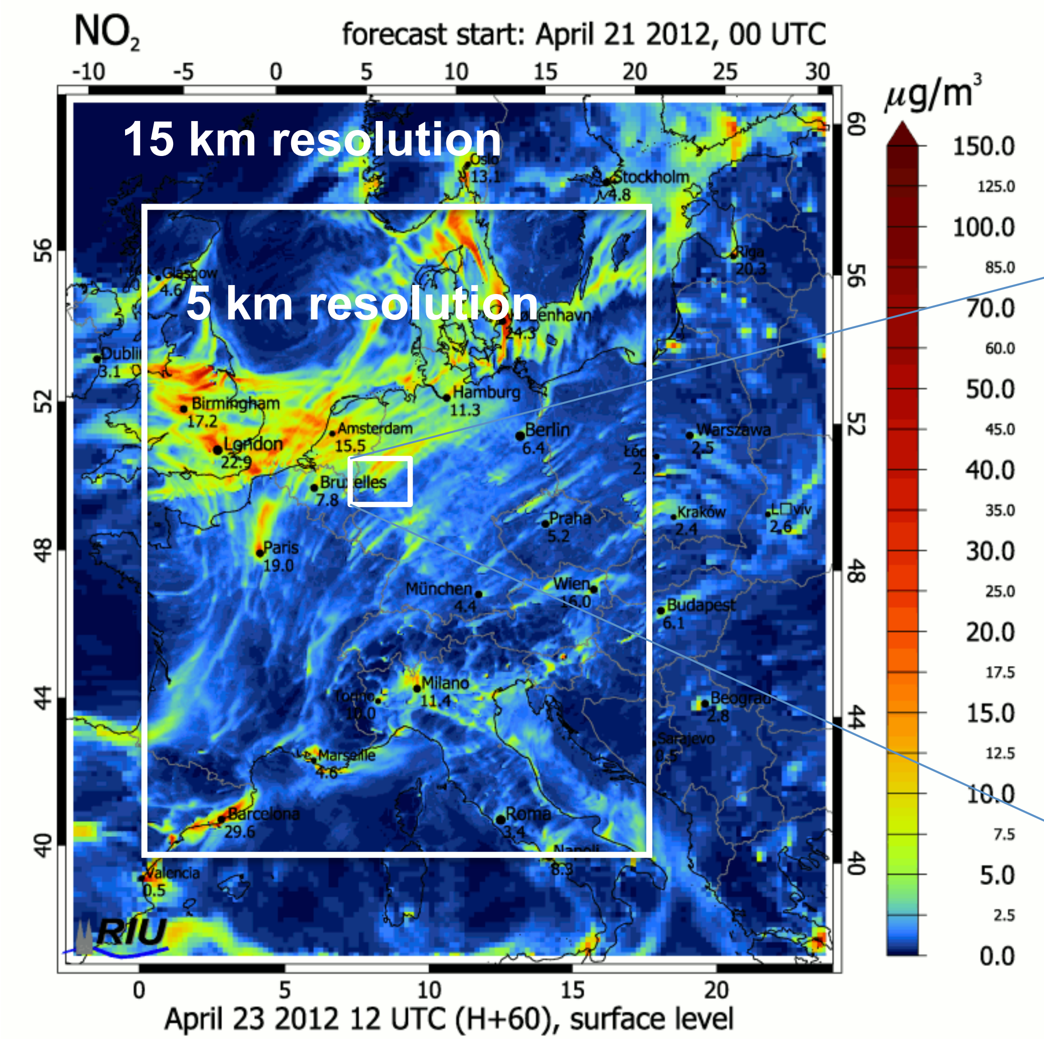
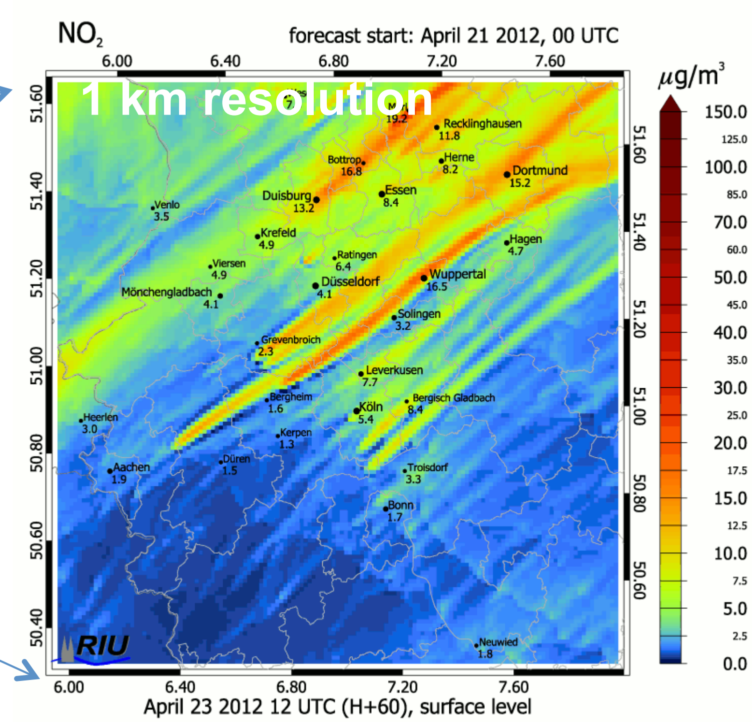
(Troposphere, IEK-8),
Research Centre Jülich, Germany

Introduction: Setting the scene

EURAD-IM nesting



EURAD-IM DS-PUBLIC Ruhr-Rur Area nested into the AIRSHED domain



44 hPa, 18.8.2002

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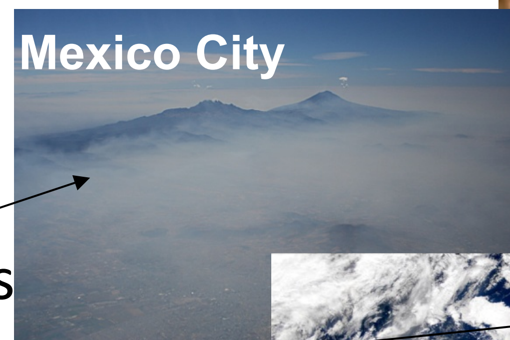
1. **Introductory remarks**
2. **Where is atmospheric chemistry a challenge for data assimilation ? (focal topics)**
   1. strongly anisotropic covariances
   2. diversity of chemical regimes: validation
   3. **varying value of observations**
   4. **more than initial values to be optimised**
3. Aerosol data assimilation

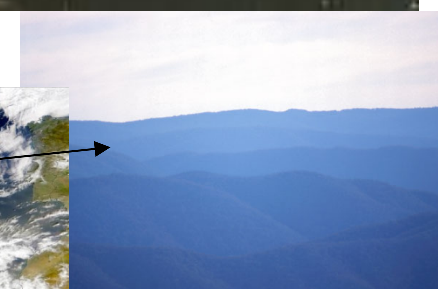
Chemistry-transport model EURAD-IM
(Reaction-advevtions-diffusion equation)
and further simulated processes

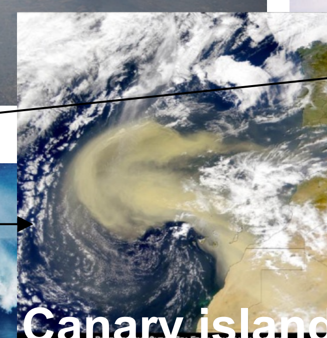
- Optionale **Chemiemechanismen,**
  ca. 60 gas phase constituents

- **aqueous phase chemistry**
  (0 ~5 species)

- **aerosols** (+ > 50 parameters
  - inorganic
  - secondary organic
  - mineral dust
  - sea salt
  - wild fires







Objectives of air quality data assimilation

- bring together air quality measurements and CTMs to provide optimal spatio-temporal reconstructions of air quality parameters,
- estimate variability, sources, sinks, and trends
- provide better air quality predictions
- reconstruct past changes,
- act as a decision support system for protection measures (which emissions are most critical?)

Which constituents/complexity really matters?

- Human health:
  - **PM_{10}**, $PM_{2.5}$, PM_1 *(= Particulate Matter)*
  - 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, other vulnerable ecosystems
  - "Acid rain"
  - ozone

$$\int_0^x m(r)dr$$

Special challenges of **tropospheric** chemistry data assimilation

The following problems prevail:

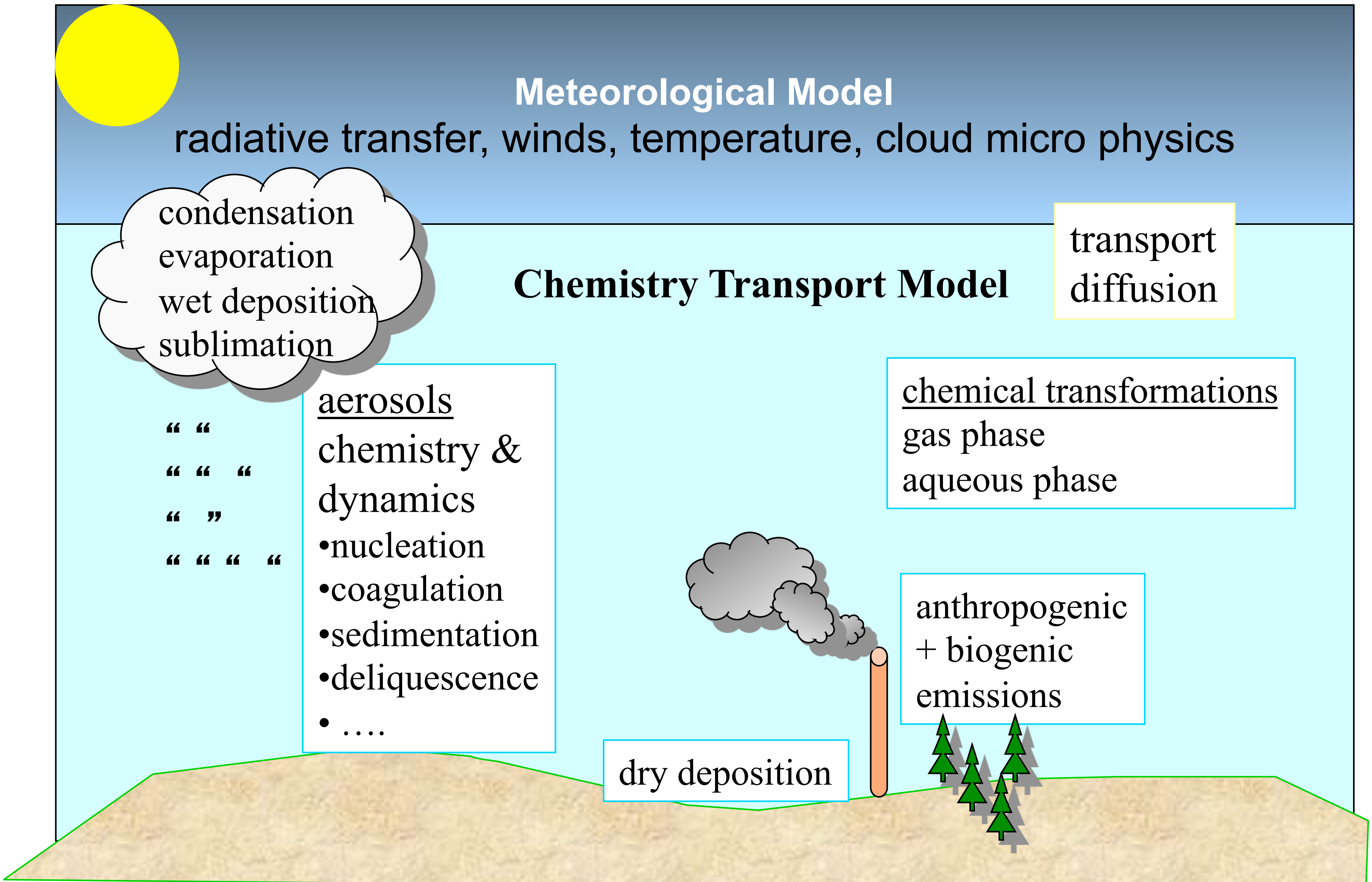
1. strong influence of manifold processes including *emissions* and *deposition*
2. *temporarily highly variable* "chemical regimes"
3. chemical state observability (= "analyseability") hampered by *manifold* ***hydrocarbon*** species
4. consistency with *heterogeneous* data sources: satellite data and in situ observations

State of the Art Examples

How are today's services designed?

1. solvers of a **deterministic** transport-diffusion-reaction equation, mostly operator-split-wise (except probably reaction-vert. diffusion)
2. continental, or regional nest ($1\ \text{km} < \Delta x < 250\ \text{km}$ horizontal resolution)
3. tropospheric gas phase chemistry, 40-100 species, < 200 reactions
4. basic aerosol treatment of at least NH_3, HNO_3, H_2SO_4

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



Special challenge: which is the appropriate scale for air quality modelling?

- intercontinental / long range transport of pollutants
- many processes are local: point and line source emissions
- surface patterns/tesselation affects simulation skills
- boundary layer and convection simulation at least of like importance as in meteorology

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3. What is the state of the art **in the troposphere?**

Some advanced auxiliary calculations (1)
singular value analysis

unit constraint (scalar product):

$$(\epsilon, \mathbf{C}\epsilon) = 1$$

maximise
Raleigh quotient:

$$\max_i \left(\frac{(\mathbf{PM}(t_1, t_0)\epsilon_i, \mathbf{EPM}(t_1, t_0)\epsilon_i)}{(\epsilon_i, \mathbf{C}\epsilon_i)} \right)$$

⇔maximise

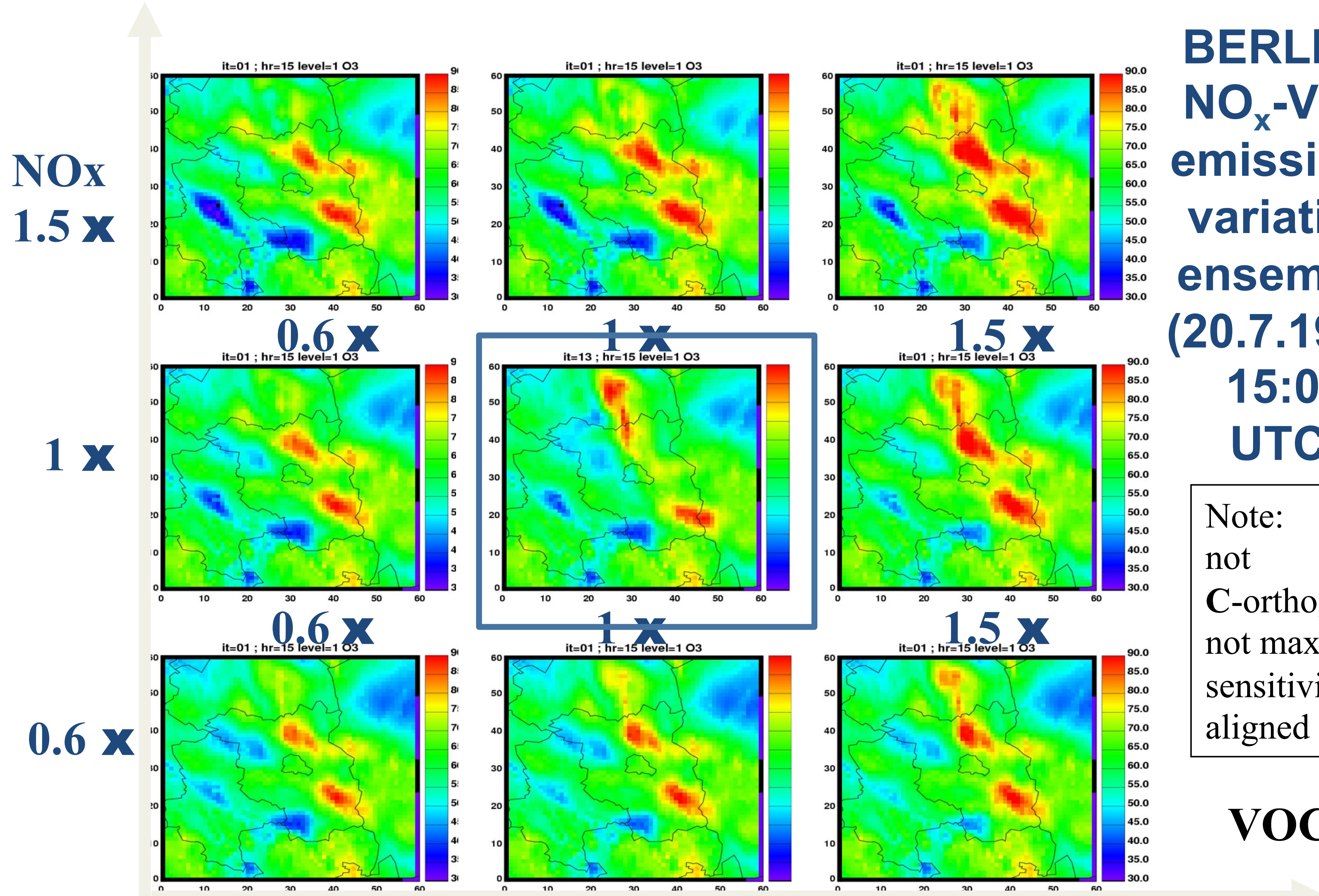
$$J(\epsilon) = (\mathbf{PM}(t_1, t_0)\epsilon_i, \mathbf{EPM}(t_1, t_0)\epsilon_i) - \lambda(\epsilon^T \mathbf{C}\epsilon - 1)$$

⇒generalised
EV problem

$$\nabla_\epsilon J(\epsilon) = 0 \equiv \mathbf{M}^T(t_1, t_0)\mathbf{P}^\mathbf{T}\mathbf{EPM}(t_1, t_0)\epsilon = \lambda\mathbf{C}\epsilon$$

$\boldsymbol{\varepsilon}_\mathbf{i}$	perturbation vector of potential optimisation parameters: initial values boundary values, emission rates, deposition velocities
C	norm inducing pos. def., sym. operator at initial time t_0 (Mahalanobis)
M	tangent linear model
E	norm inducing pos. def., sym. operator at optimisation time t_1
(P	projection operator, extinguishing areas or species outside focus)
λ	Lagrange parameter and generalised eigenvalues

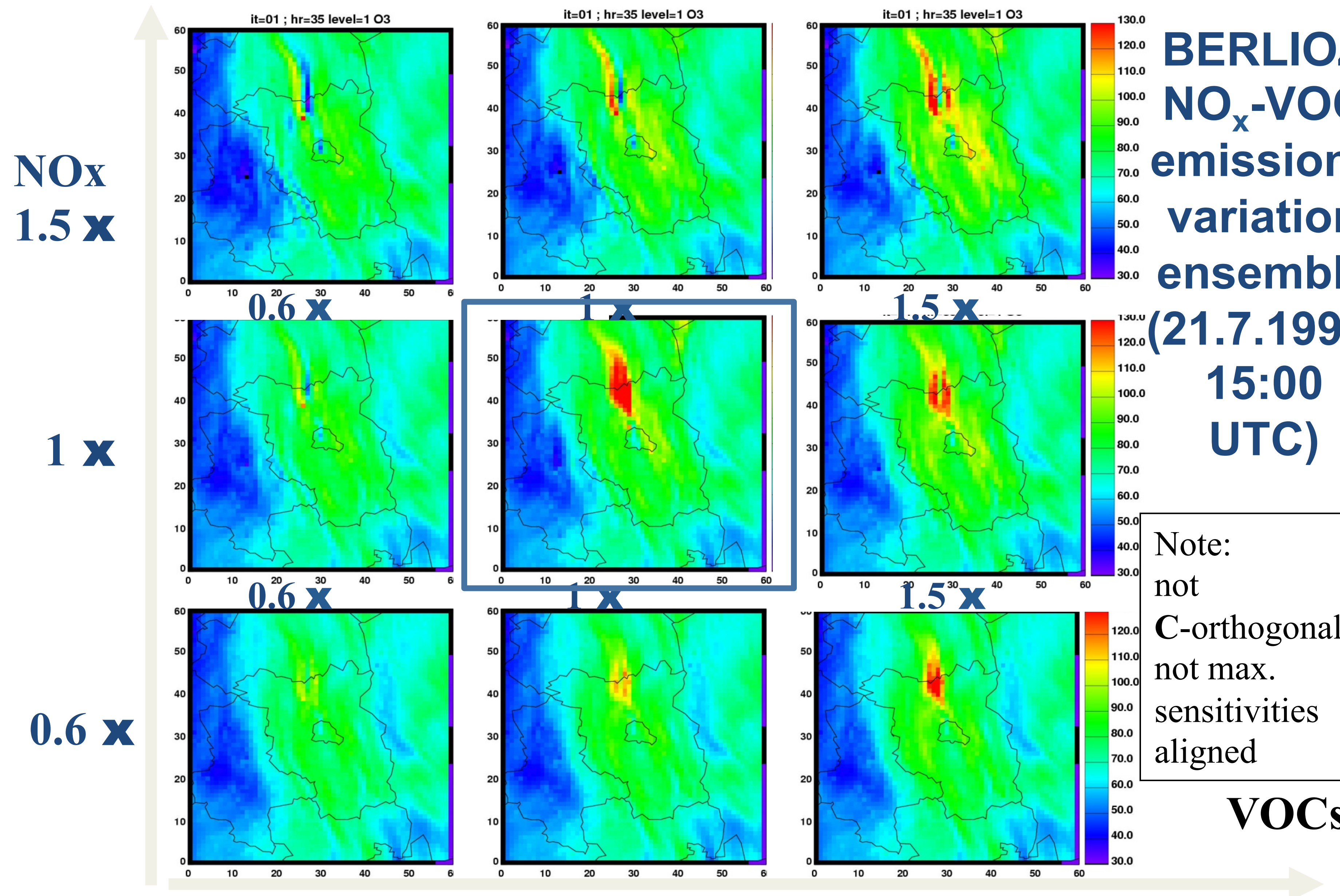
3. Focus: varying value of observations



BERLIOZ NO_x-VOC emissions variation ensemble (20.7.1998, 15:00 UTC)

Note:
not **C**-orthogonal,
not max. sensitivities aligned

3. Focus: varying value of observations



BERLIOZ NO_x-VOC emissions variation ensemble (21.7.1998, 15:00 UTC)

Note: not **C**-orthogonal, not max. sensitivities aligned

BLUE-based assimilation algorithms

$$J(\mathbf{x}) = \frac{1}{2}[\mathbf{x}^b - \mathbf{x}]^T \mathbf{B}_0^{-1}[\mathbf{x}^b - \mathbf{x}] \; + \frac{1}{2}\left\{\mathbf{y}^0 - H[\mathbf{x}(t)]\right\}^T \mathbf{R}^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x}]\right\}$$

The gradient then reads

$$\nabla J(\mathbf{x}) = \mathbf{B}_0^{-1}[\mathbf{x}^b - \mathbf{x}] \; + H^T \mathbf{R}^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x} + (\mathbf{x}^b - \mathbf{x}^b)]\right\}$$

where a trivial expansion is introduced for later manipulation.

At the minimum (=optimum, = analysis, $\mathbf{x} = \mathbf{x}_a$) $\nabla J(\mathbf{x}_a) = 0$

$$\nabla J(\mathbf{x}) = 0 = \mathbf{B}_0^{-1}[\mathbf{x}^b - \mathbf{x}_a] \; + H^T \mathbf{R}^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x}^b]\right\} \; - H^T \mathbf{R}^{-1} H[\mathbf{x}_a - \mathbf{x}^b],$$

from which follows:

$$\mathbf{x}_a - \mathbf{x}_b = (\mathbf{B}_0^{-1} + \mathbf{H}^T \mathbf{R}^{-1}\mathbf{H})^{-1}\mathbf{H}^T \mathbf{R}^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x}^b]\right\}$$

$$= \mathbf{B}\mathbf{H}^T(\mathbf{R} + \mathbf{H}^T\mathbf{B}\mathbf{H})^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x}^b]\right\}$$

with the latter result obtained after some manipulation.

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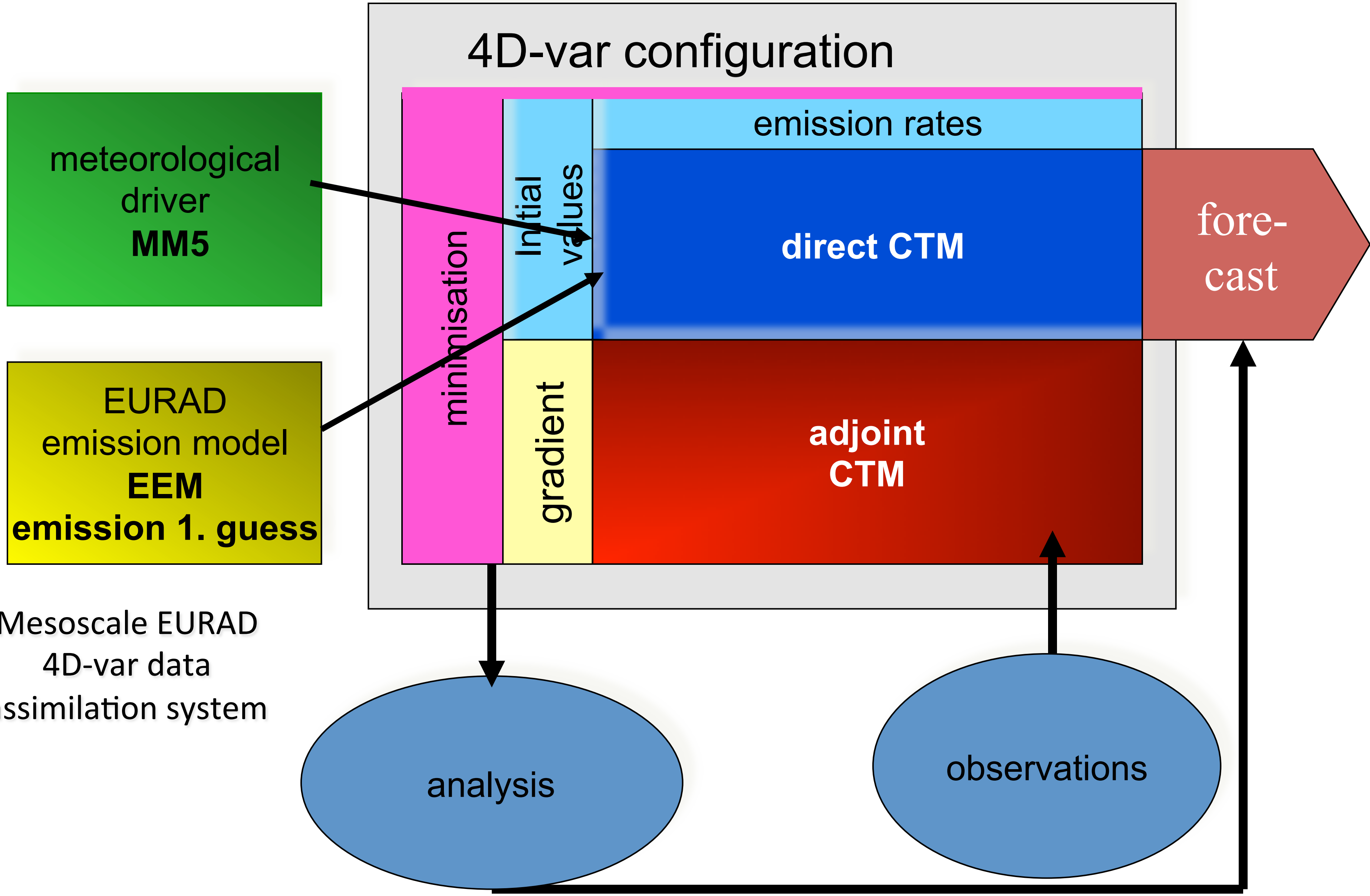
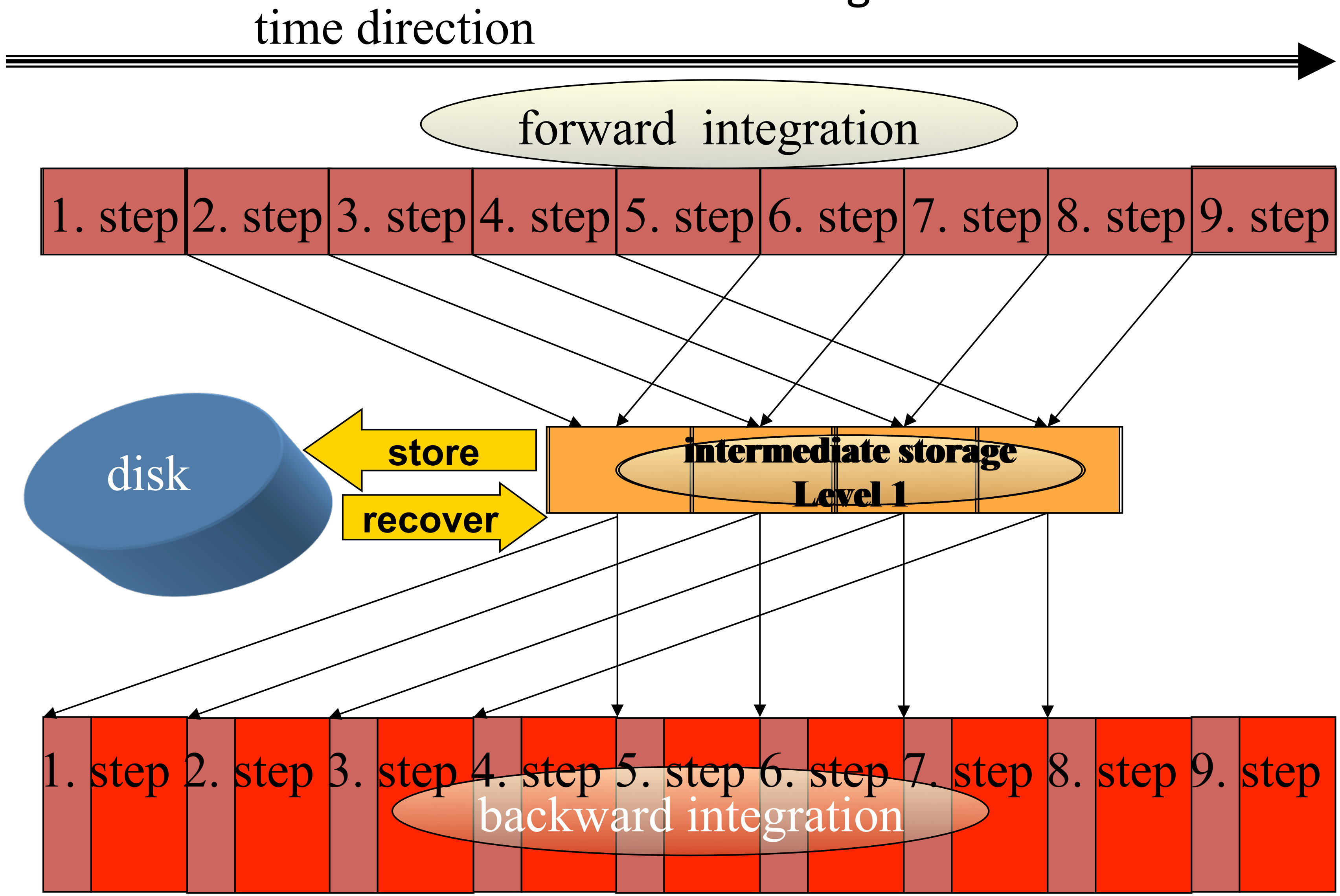
Components of a chemistry 4D-var system



Mesoscale EURAD
4D-var data
assimilation system

4. Focus: joint emission rate initial value optimisation

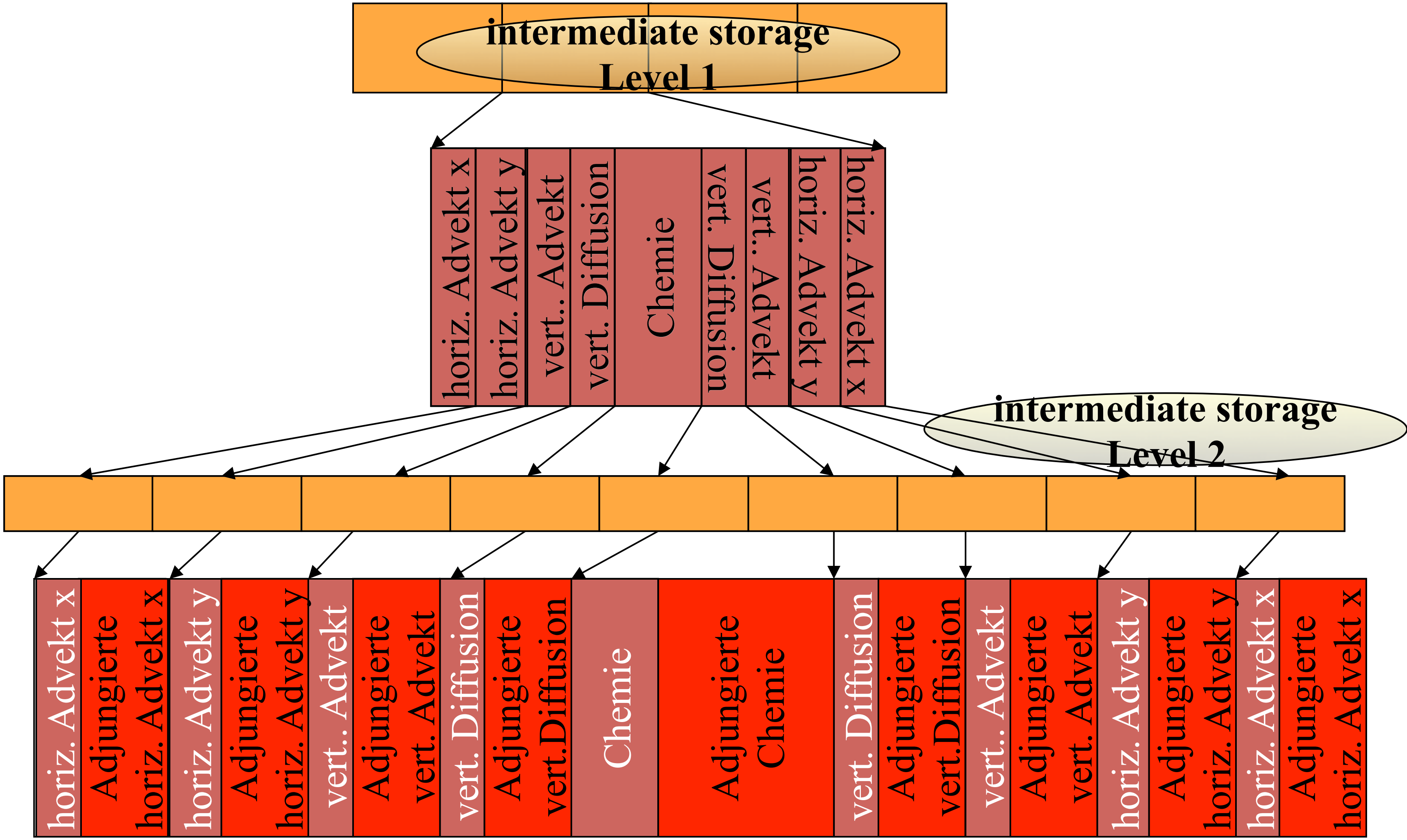
2 level forward and backward integration scheme



4. Focus: joint emission rate initial value optimisation

storage sequence: level 2, operator split

(each time step)



Advanced spatio-temporal methods used in tropospheric chemistry data assimilation

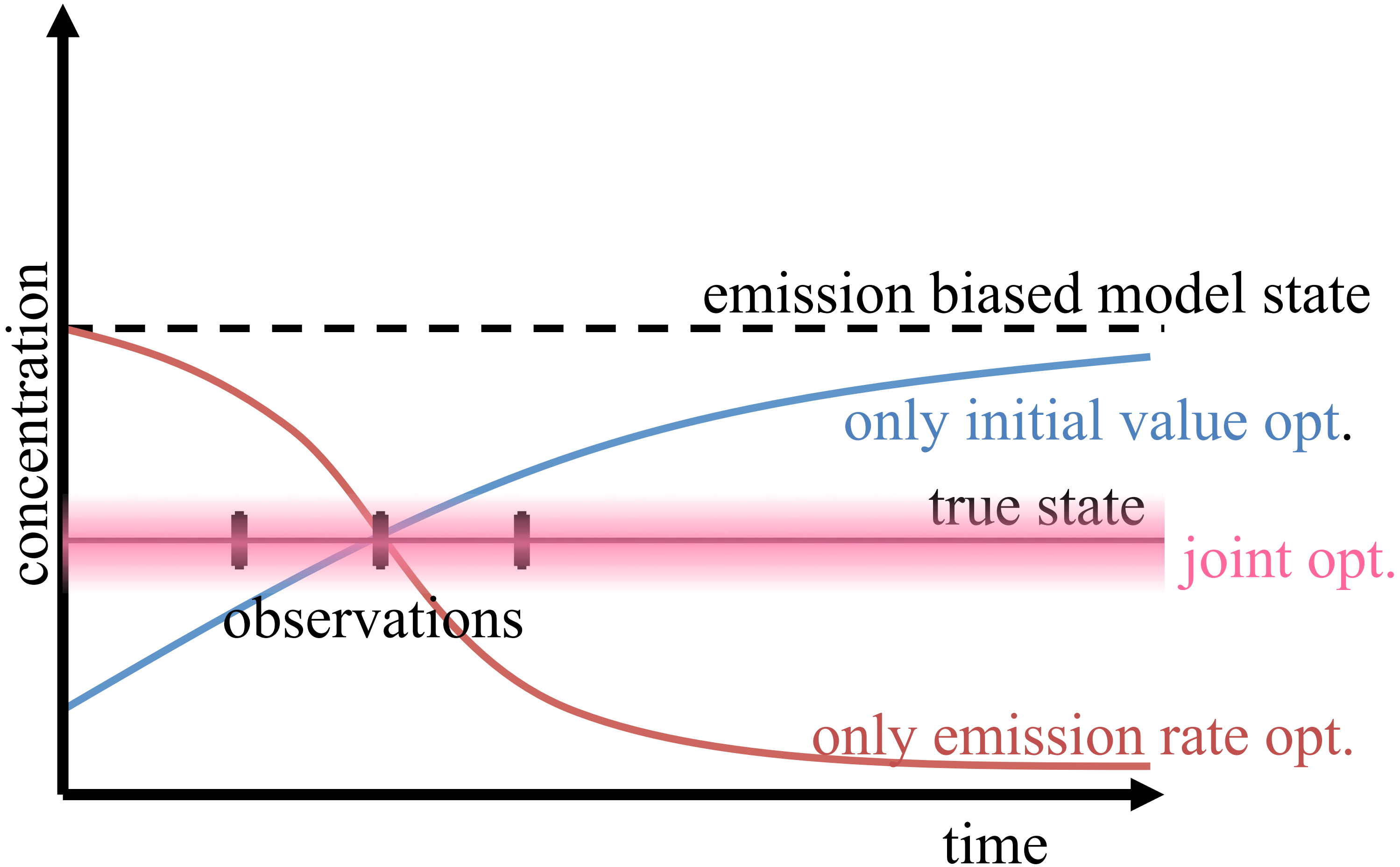
Spacio-temporal BLUEs applied in tropospheric chemistry data assimilation:

- 4D var:
  - with EURAD (Elbern and Schmidt, 1999, 2001),
  - with POLAIR (Issartel and Baverel, 2003)
- Kalman Filter
  - with LOTOS model (van Loon et al, 2000), (RRSQR)
  - with EUROS model (Hanea et al. 2004) (En+RRSQRKF)

Question: Which parameter to be optimized?

Hypothesis:

initial state and emission rates are least known



In the troposphere, for **emission rates,** the product (*paucity of knowledge*importance*) is high

Emission Rate Optimization

minimize cost function

$$J(\mathbf{x}(t_0), \mathbf{e}) = \frac{1}{2}(\mathbf{x}^b(t_0) - \mathbf{x}(t_0))^T \mathbf{B}_0^{-1}(\mathbf{x}^b(t_0) - \mathbf{x}(t_0)) + \frac{1}{2}\int_{t_0}^{t_N}(\mathbf{e}_b(t) - \mathbf{e}(t))^T \mathbf{K}^{-1}(\mathbf{e}_b(t) - \mathbf{e}(t))dt + \frac{1}{2}\int_{t_0}^{t_N}\left(\mathbf{y}^0(t) - H[\mathbf{x}(t)]\right)^T \mathbf{R}^{-1}(\mathbf{y}^0(t) - H[\mathbf{x}(t)])dt$$

deviations from background initial state

deviations from a priori emission rates

model deviations from observations

$\mathbf{x}^b(t_0)$	background state at $t = 0$
$\mathbf{x}(t)$	model state at time t
$\mathbf{e}_b(t_0)$	background emission rate at $t = 0$
$\mathbf{e}(t)$	emission rate field at time t
$\mathbf{K}$	emission rate error covariance matrix
$H[\ \ ]$	forward interpolator
$\mathbf{y}^0(t)$	observation at time t
$\mathbf{B}_0$	background error covariance matrix

Incremental Formulation

- Analysis State:

$$\boldsymbol{x}^a = \boldsymbol{x}^b + \delta\boldsymbol{x}^a$$
$$\boldsymbol{u}^a = \boldsymbol{u}^b + \delta\boldsymbol{u}^a$$

- New „State“ Variables:

$$\boldsymbol{v} = \mathbf{B}^{-1/2}\delta\boldsymbol{x}$$
$$\boldsymbol{w} = \mathbf{K}^{-1/2}\delta\boldsymbol{u}$$

- Cost Function:

$$J(\boldsymbol{v},\boldsymbol{w}) = \frac{1}{2}\boldsymbol{v}^T\boldsymbol{v} + \frac{1}{2}\boldsymbol{w}^T\boldsymbol{w} + \frac{1}{2}\left[\mathbf{H}\delta\boldsymbol{x}_i - \boldsymbol{d}_i\right]^T\mathbf{R}^{-1}\left[\mathbf{H}\delta\boldsymbol{x}_i - \boldsymbol{d}_i\right]$$

- Gradient:

$$\nabla_{\boldsymbol{v}}J = \nabla_{\boldsymbol{v}}J_{IV} + \nabla_{\boldsymbol{v}}J_O = \boldsymbol{v} + \mathbf{B}^{\mathrm{T}/2}\nabla_{\delta\boldsymbol{v}}J_O$$
$$\nabla_{\boldsymbol{w}}J = \nabla_{\boldsymbol{w}}J_{EF} + \nabla_{\boldsymbol{w}}J_O = \boldsymbol{w} + \mathbf{K}^{\mathrm{T}/2}\nabla_{\delta\boldsymbol{w}}J_O.$$

Background Error Covariance Matrix B

- must be provided as an operator (size is of order 10^{13})
- we would like to have an operator which can easily be factorised by $B=B^{1/2}B^{T/2}$
- *Weaver and Courtier (2001):*
  - general diffusion equation serves for a valid operator generating a positive definite covariance operator
  - diffusion equation is self adjoint
  - $B^{1/2}$ and $B^{T/2}$ by applying the diffusion operator half the diffusion time

Transport-diffusion-reaction equation and its adjoint

Tendency Equations

direct chemistry transport equation

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

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

$$-\frac{\partial \delta c_i^*}{\partial t} - \mathbf{v} \nabla \delta c_i^* - \frac{1}{\rho} \nabla \cdot (\rho \mathbf{K} \nabla \delta c_i^*) + \Sigma_{r=1}^{R} \left(k(r) \frac{s_i(r_-)}{c_i} \Pi_{j=1}^{U} \bar{c}_j^{\,s_j(r_-)} \Sigma_{n=1}^{U} \left(s_n(r_+) - s_n(r_-) \right) \delta c_n^* \right) = 0$$

Adjoint integration "backward in time"

direct model

$$\frac{d\mathbf{x}}{dt} = \mathcal{M}(\mathbf{x})$$

tangent linear model

$$\delta\mathbf{x}(t_n) = \mathbf{M}'(t_n, t_0)\delta\mathbf{x}(t_0) = \prod_{i=n}^{1} \mathbf{M}'(t_i, t_{i-1})\delta\mathbf{x}(t_0)$$

adjoint model

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

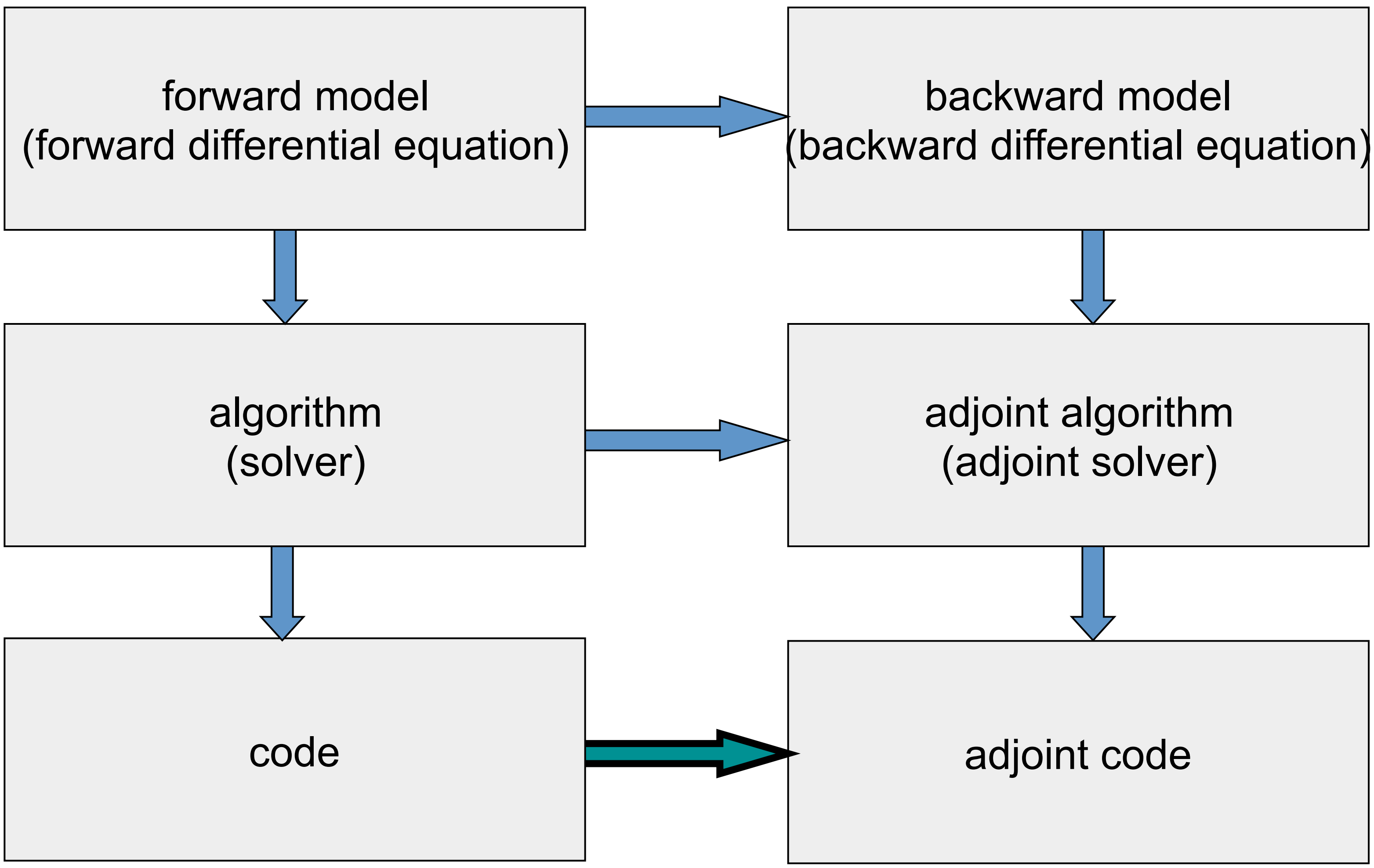
$$\nabla_{[\mathbf{x}(t_0),\mathbf{e}]}J = -\mathbf{B}_0^{-1}(\mathbf{x}^b(t_0) - \mathbf{x}(t_0)) - \mathbf{K}^{-1}(\mathbf{e}^b(t) - \mathbf{e}(t)) - \Sigma_{m=0}^{N}\Pi_{i=1}^{m}\mathbf{M}^T(t_{i-1}, t_i)\mathbf{R}^{-1}(\mathbf{y}^0(t_m) - H[\mathbf{x}(t_m)])$$

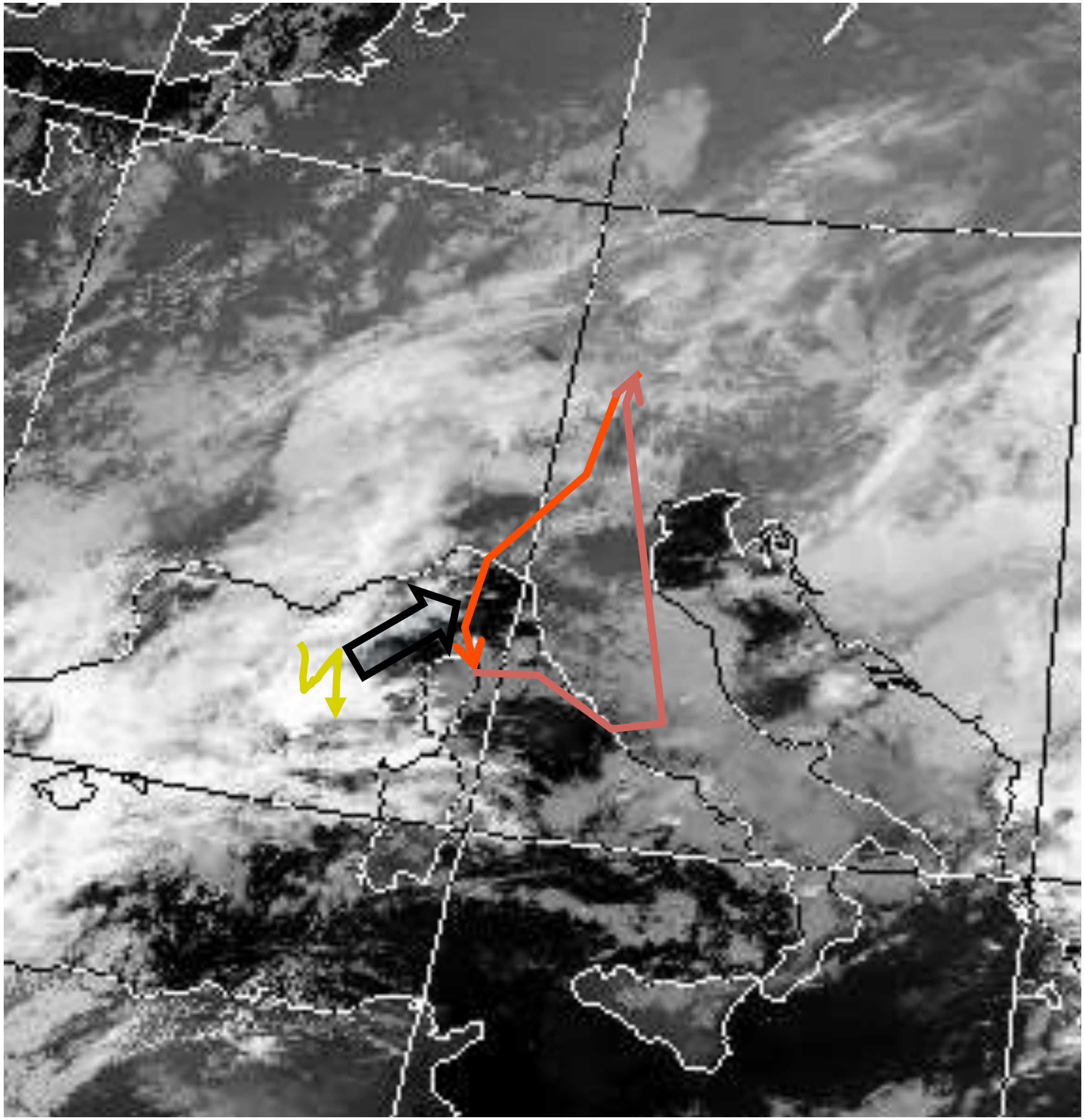
Find minimum of $J(\mathbf{x}(t_0), \mathbf{e})$ with $\nabla_{[\mathbf{x}(t_0),\mathbf{e}]}J$ by use of a minimization routine

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

back

The 4D variational method:
Key development: construction of the adjoint code





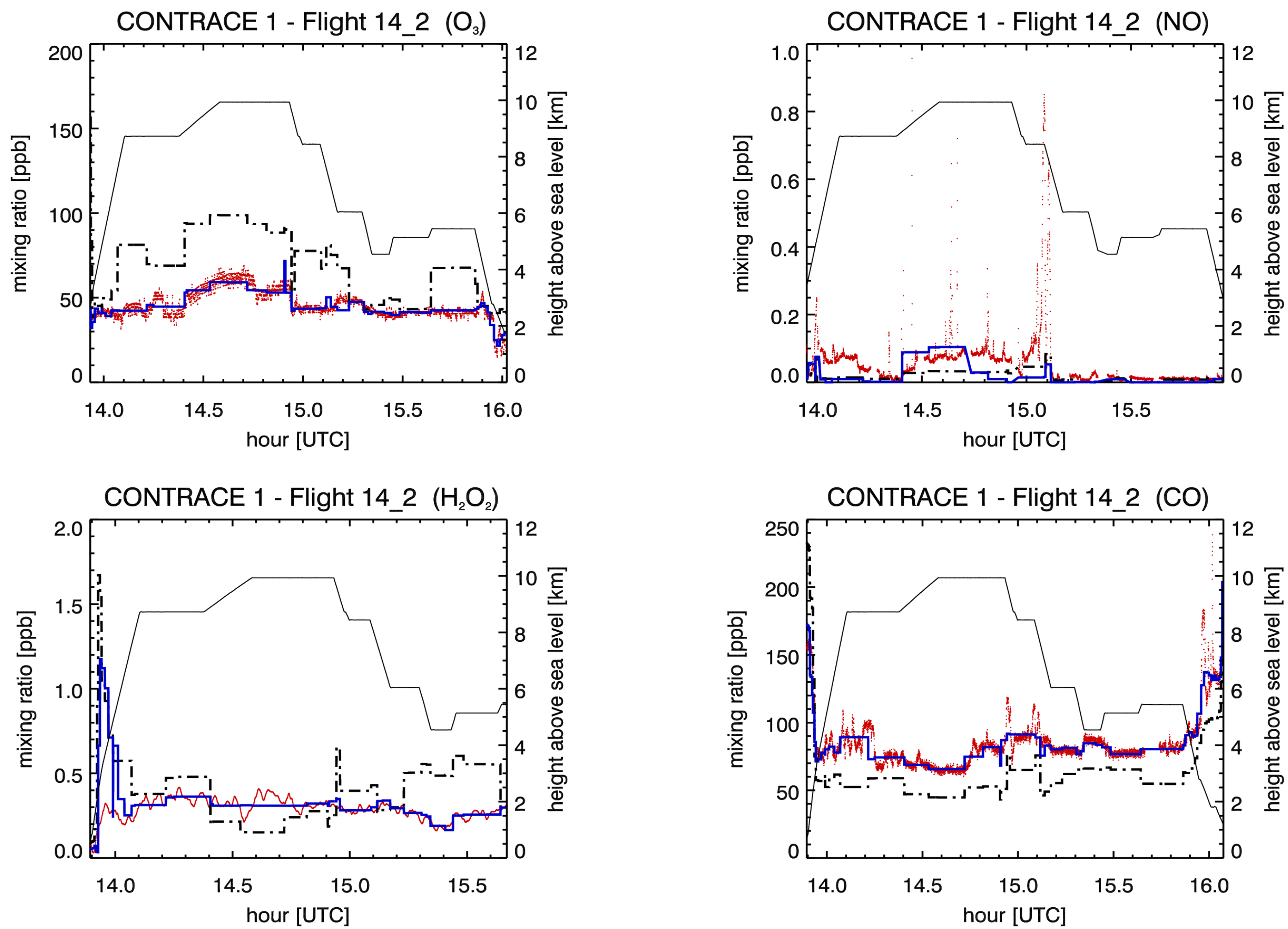
CONTRACE
Convective Transport of Trace Gases into the upper Troposphere over Europe: Budget and Impact of Chemistry

Coord.: *H. Huntrieser, DLR*

flight path Nov. 14, 2001

CONTRACE
Nov. 14, 2001 north (= home) bound

O_3 NO H_2O_2 CO



1. guess — assimilation result — observations — flight height [km]

Kalman filter, basic equations

forecast

$$\mathbf{x}^f(t_i) = \mathbf{M}(t_i, t_{i-1})\mathbf{x}^a(t_{i-1}) + \eta \tag{1}$$

background error covariance matrix

$$\mathbf{P}_i^b = \mathbf{M}(t_i, t_{i-1})\mathbf{P}_i^a\mathbf{M}^T(t_i, t_{i-1}) + \mathbf{Q}$$

optimally estimated state

$$\mathbf{x}^a(t_i) = \mathbf{x}^b(t_i) + \mathbf{K}_i\mathbf{d}_i, \tag{1}$$

Kalman gain matrix

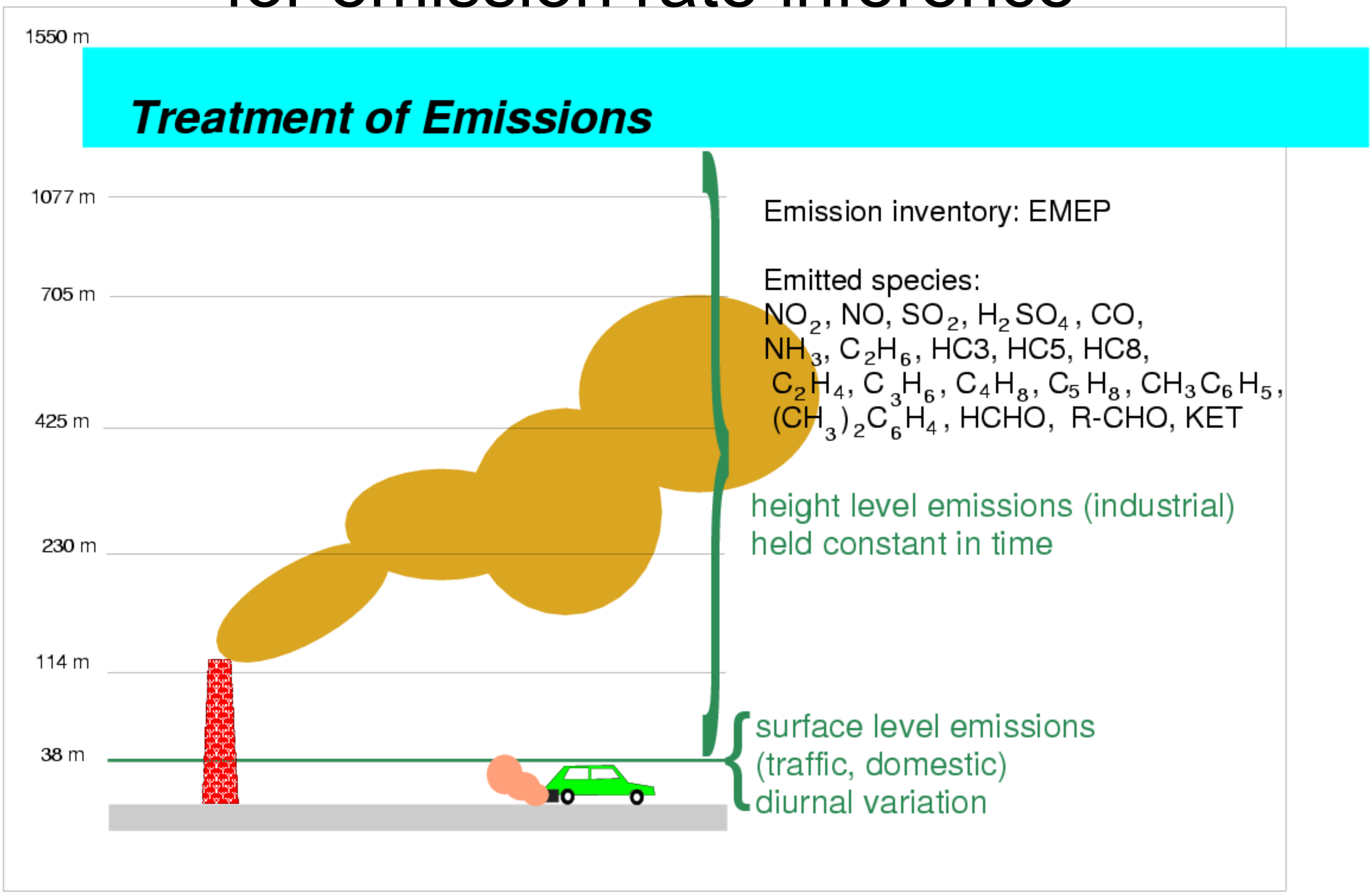
$$\mathbf{K}_i := \mathbf{P}_i^b\mathbf{H}_i^T(\mathbf{H}_i\mathbf{P}_i^b\mathbf{H}_i^T + \mathbf{R}_i)^{-1} \quad \in \mathcal{R}^{n\times p_i} \tag{2}$$

and analysis error covariance matrix

$$\mathbf{P}_i^a = (\mathbf{I} - \mathbf{K}_i\mathbf{H})\mathbf{P}_i^b. \tag{3}$$

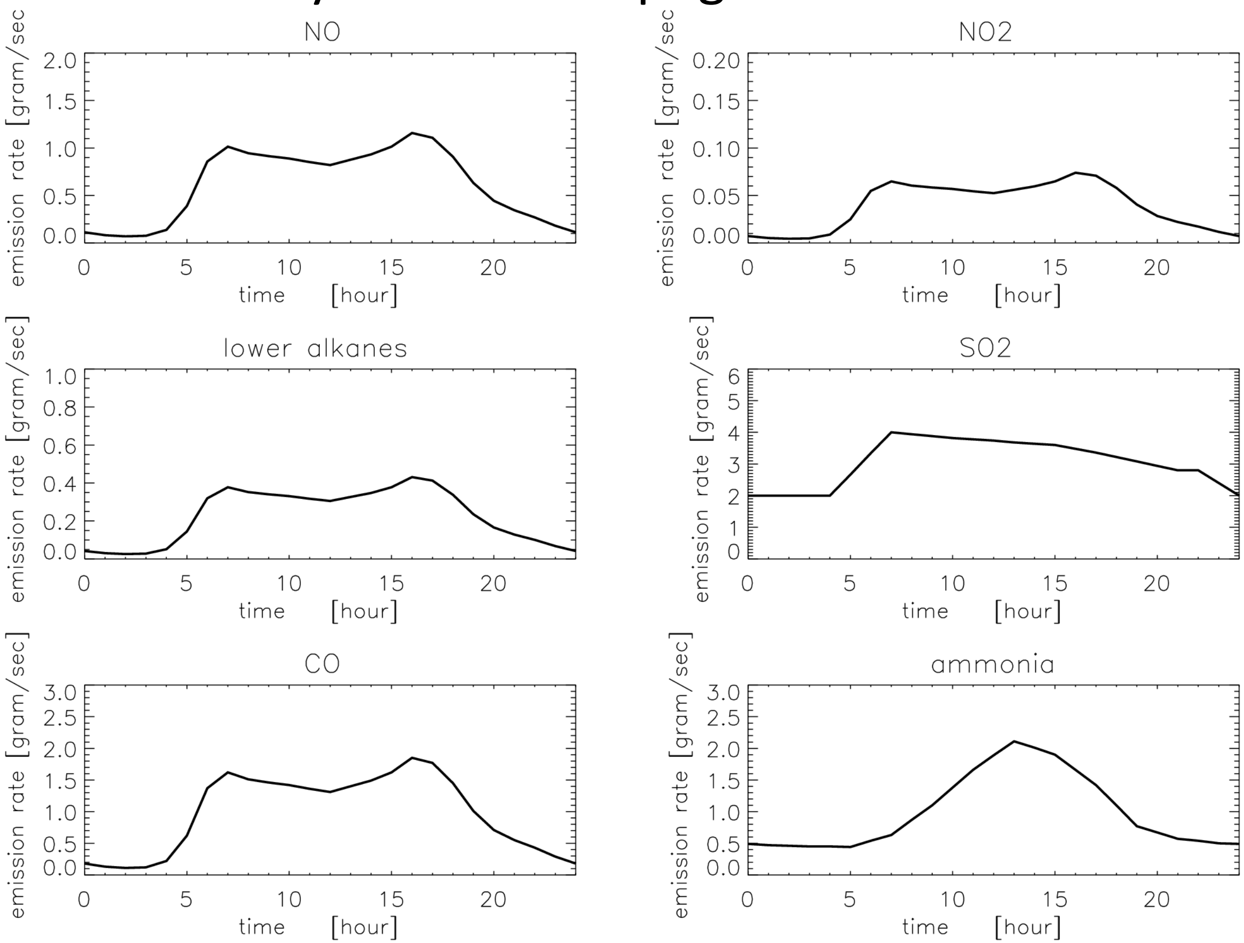
4. Focus: joint emission rate initial value optimisation

Treatment of the inverse problem for emission rate inference



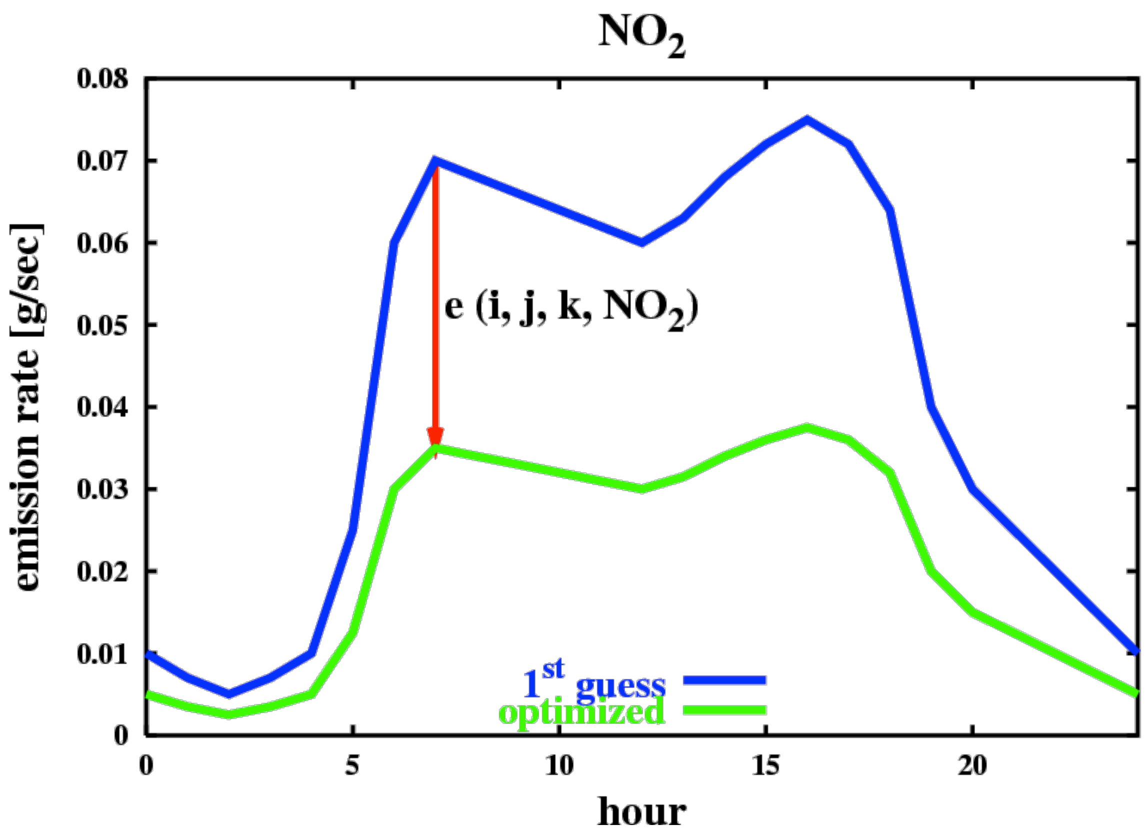
4. Focus: joint emission rate initial value optimisation

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



Optimisation of emission rates

amplitude optimisation for each emitted species and grid cell

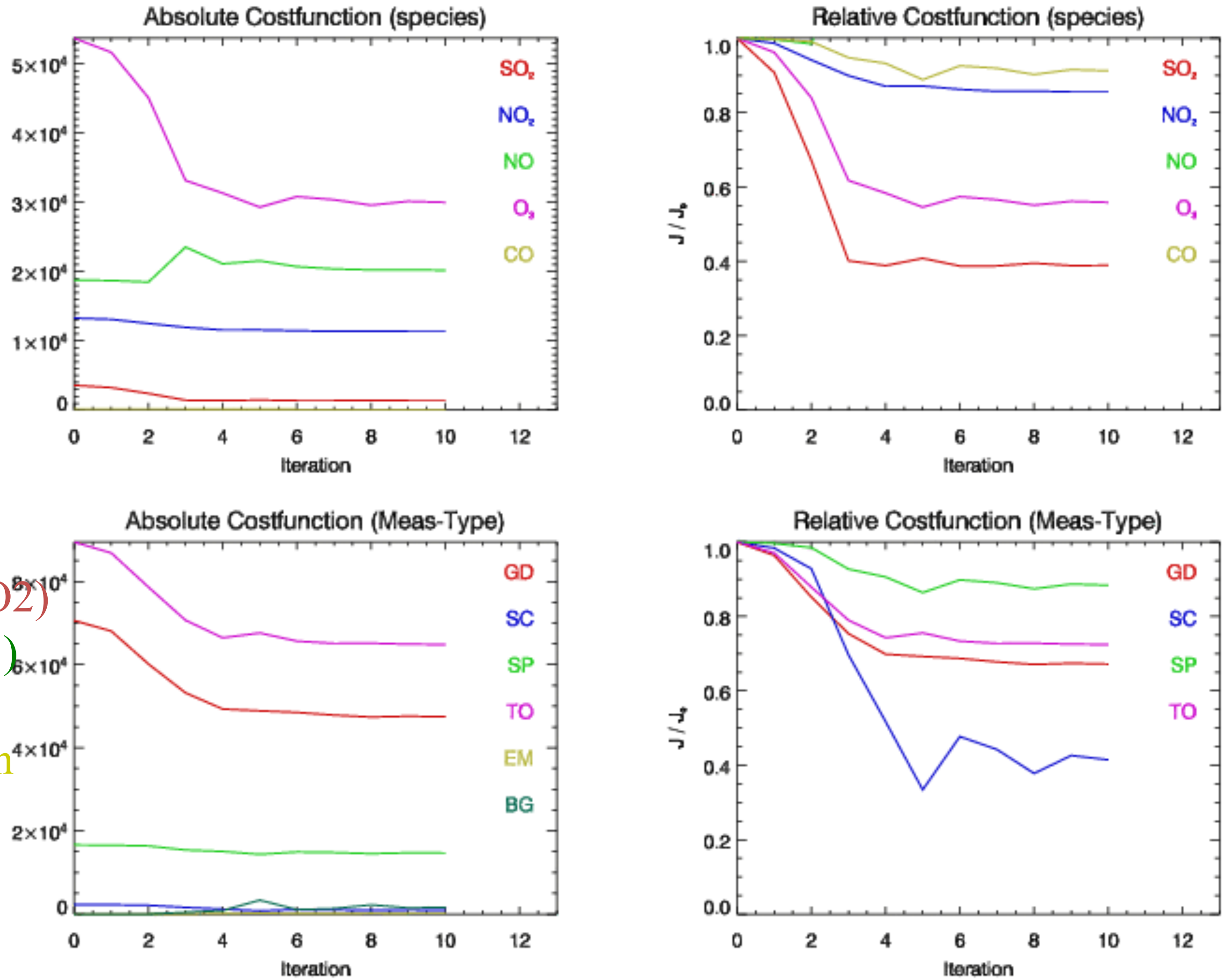


Background emission rate covariance matrix D

	SO2	SO4	NO2	NO	ALD	HCHO	NH3	HC3	HC5	HC8	ETH	CO	OL2	OLT	OLI	TOL	XYL	KET	ISO
SO2	100	40	12	12	0.4	0.4	0.4	0.4	0.4	0.4	0.4	9	0.4	0.4	0.4	0.4	0.4	0.4	0
SO4		100	12	12	0.4	0.4	0.4	0.4	0.4	0.4	0.4	9	0.4	0.4	0.4	0.4	0.4	0.4	0
NO2			100	26	8	8	7	8	8	8	8	12	8	8	8	7	7	8	0.4
NO				100	8	8	7	8	8	8	8	12	8	8	8	7	7	8	0.4
ALD					100	26	5	6	6	6	6	5	10	6	6	5	5	26	0.3
HCHO						100	5	6	6	6	6	5	10	6	6	5	5	26	0.3
NH3							100	5	5	5	5	5	5	5	5	5	5	5	0.3
HC3								100	23	23	23	5	18	15	15	6	6	6	0.3
HC5									100	23	23	5	18	15	15	6	6	6	0.8
HC8										100	23	5	18	15	15	6	6	6	0.8
ETH											100	5	18	15	15	6	6	6	0.8
CO												100	6	5	5	5	5	5	0.3
OL2													100	23	23	6	6	10	1
OLT														100	27	5	5	6	15
OLI															100	5	5	6	15
TOL																100	24	5	0.3
XYL																	100	5	0.3
KET																		100	0.3
ISO																			100

- > 10 %
- > 1 %
- < 1 %

Reduction of the partial cost functions relative to observation type (coarse grid, 54 km)



GO ground in situ

SC satellite columns (NO2)

SP satellite profiles (O3)

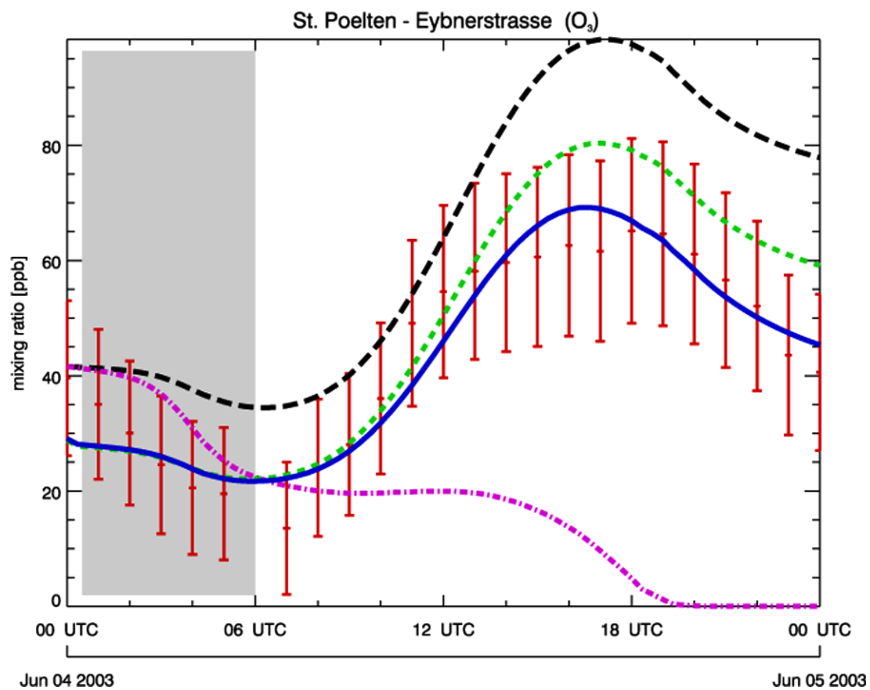
TO total costs

EM background emission

BG state background

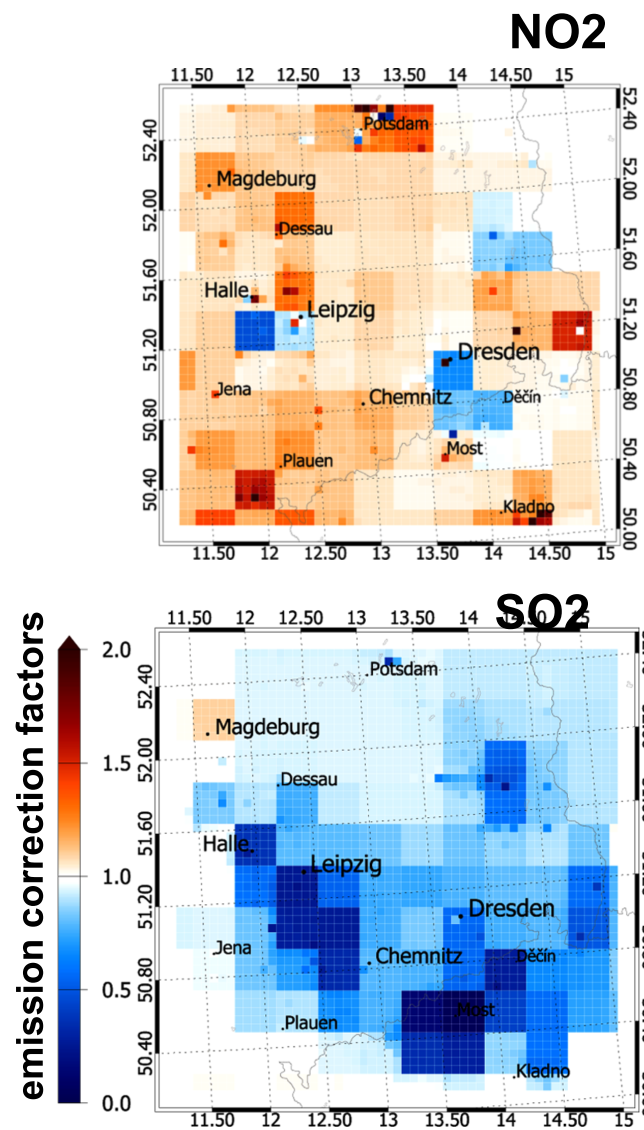
EURAD IM System

2. Analyses, Example (i): Analysis of emissions by 4D-var (VERTIKO)

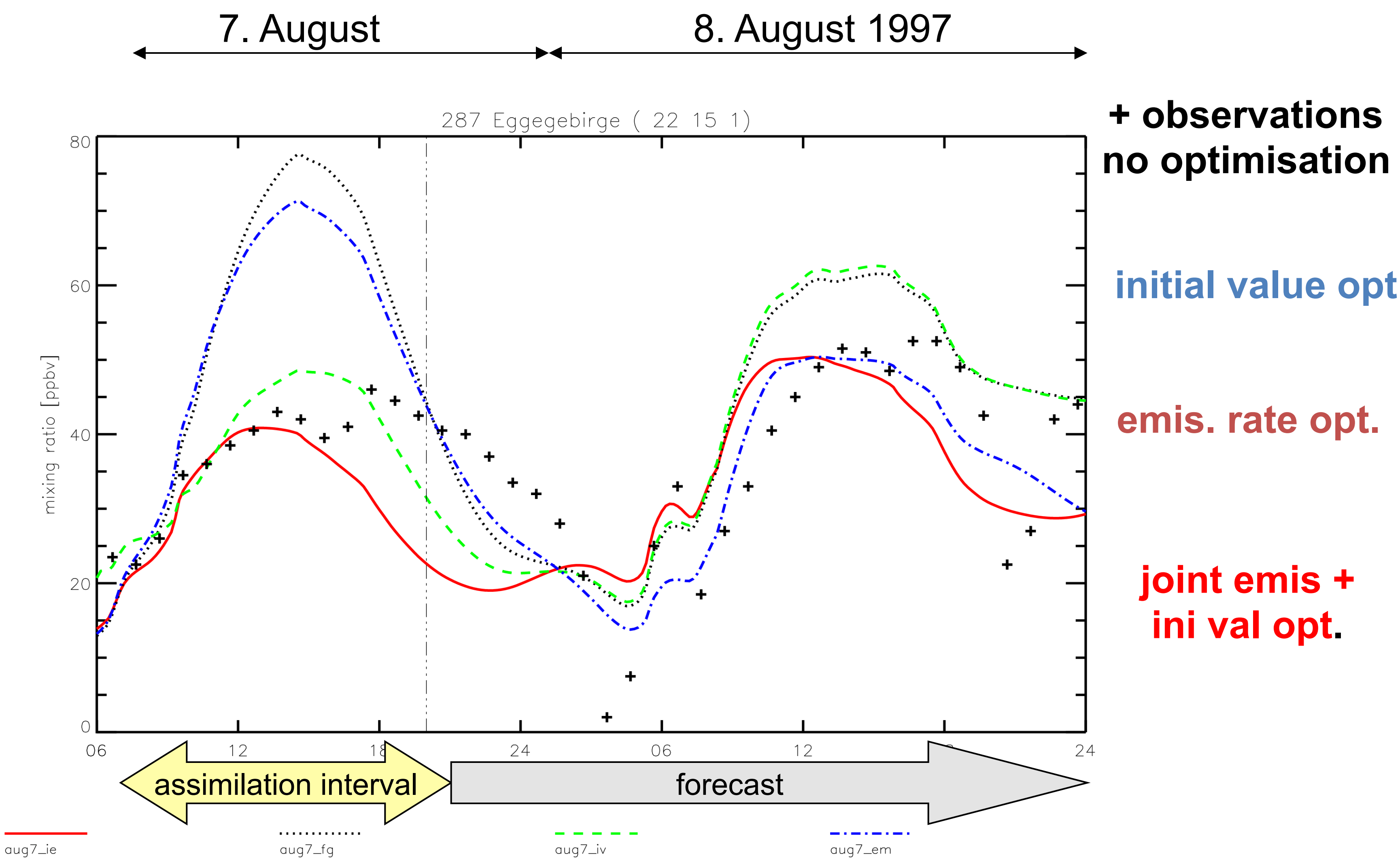


Observed and analysed ozone evolution at St. Poelten Vertical bars: ozone observations with error estimates.

- - - - - Control run without data assimilation.
...... initial value optimisation.
-.-.-.-.-. emission factor optimisation.
——— joint initial value and emission factor optimisation (Strunk et al., 2011)

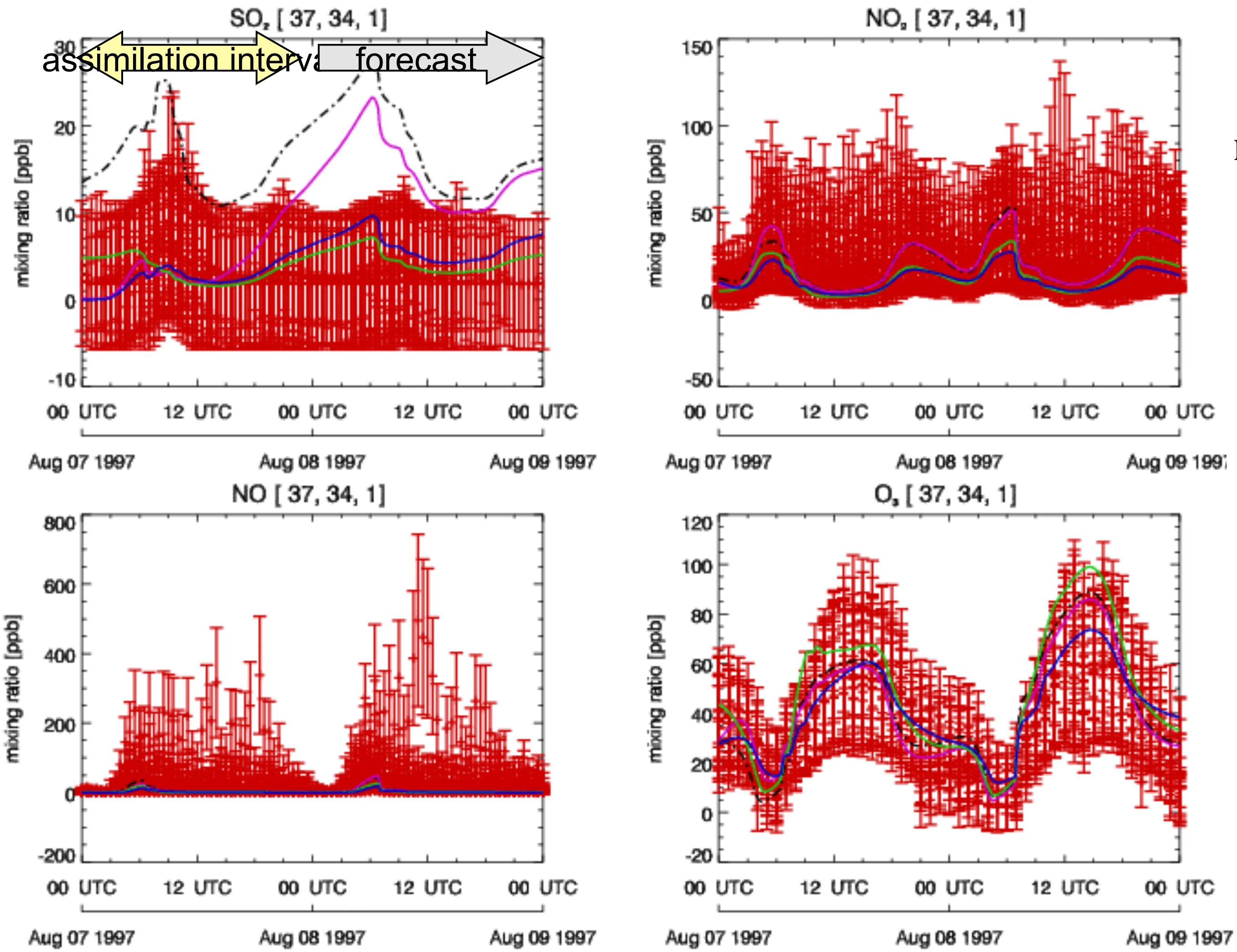


Semi-rural measurement site **Eggegebirge**



4. Focus: joint emission rate initial value optimisation

Rhine-Main area box: (Frankfurt-Mainz)
9.-10. August 1997



+ observations
no optimisation

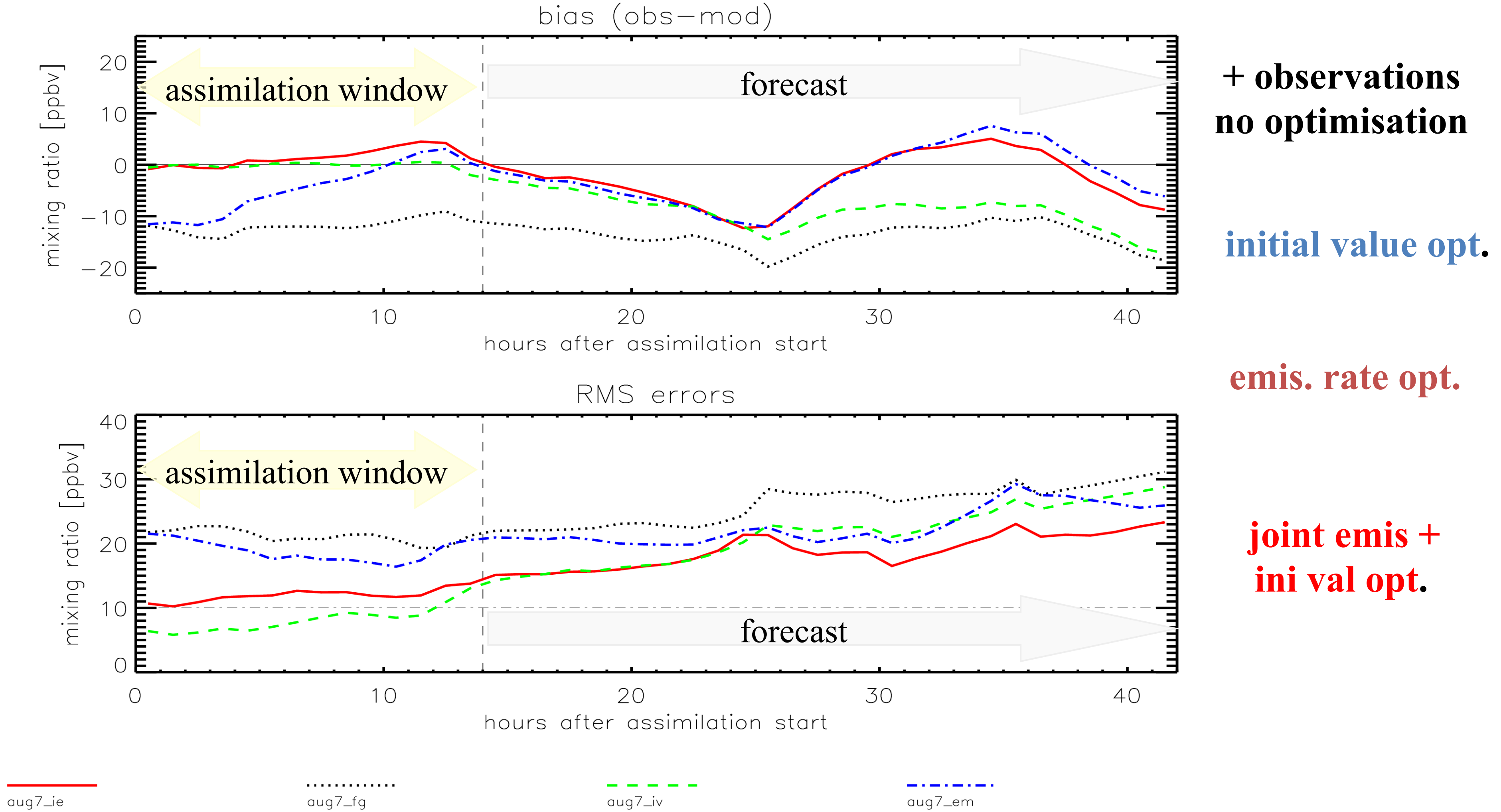
initial value opt.

emis. rate opt.

joint emis + ini val opt.

4. Focus: joint emission rate initial value optimisation

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

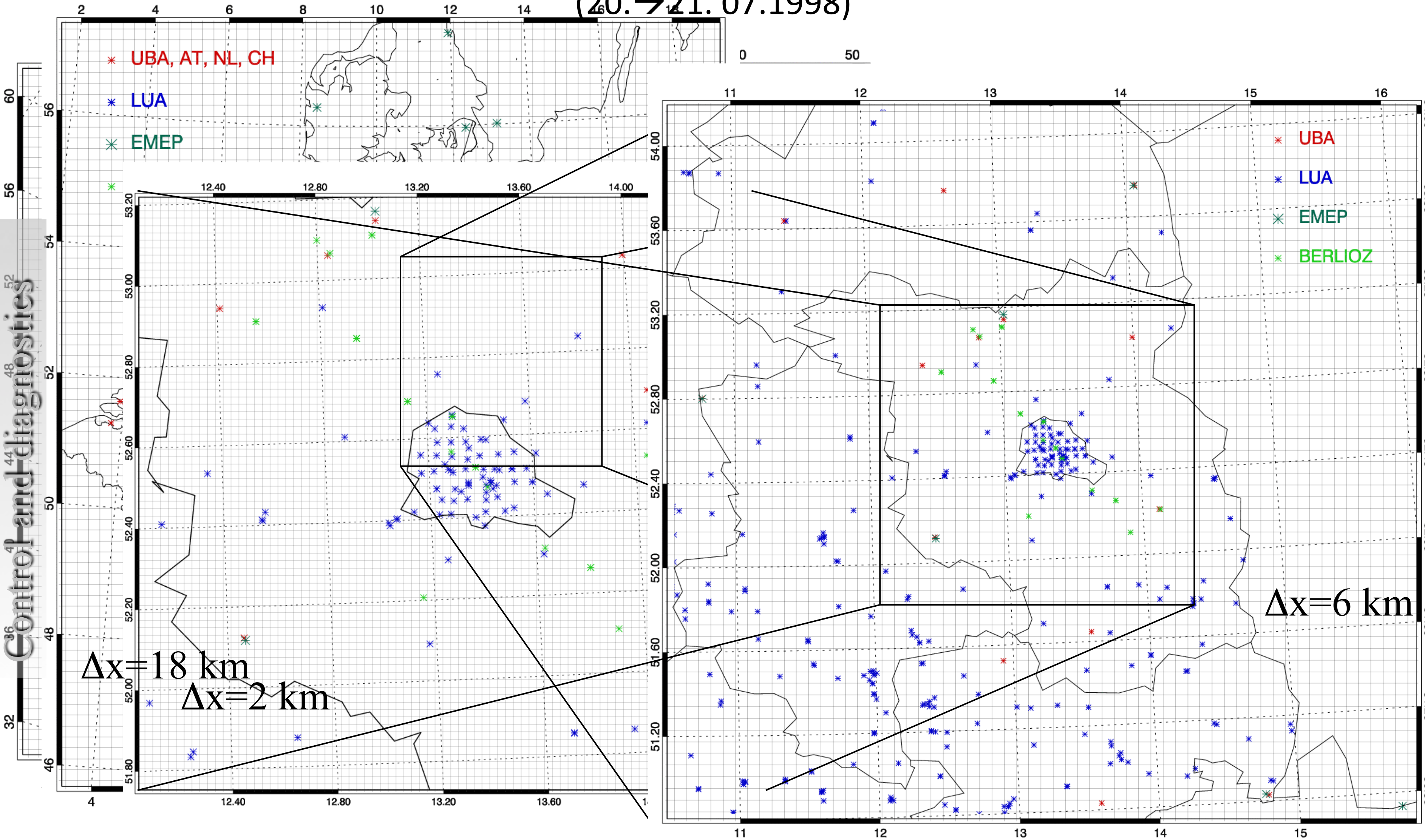


4. Focus: joint emission rate initial value optimisation

Which is the requested resolution?

BERLIOZ grid designs and observational sites

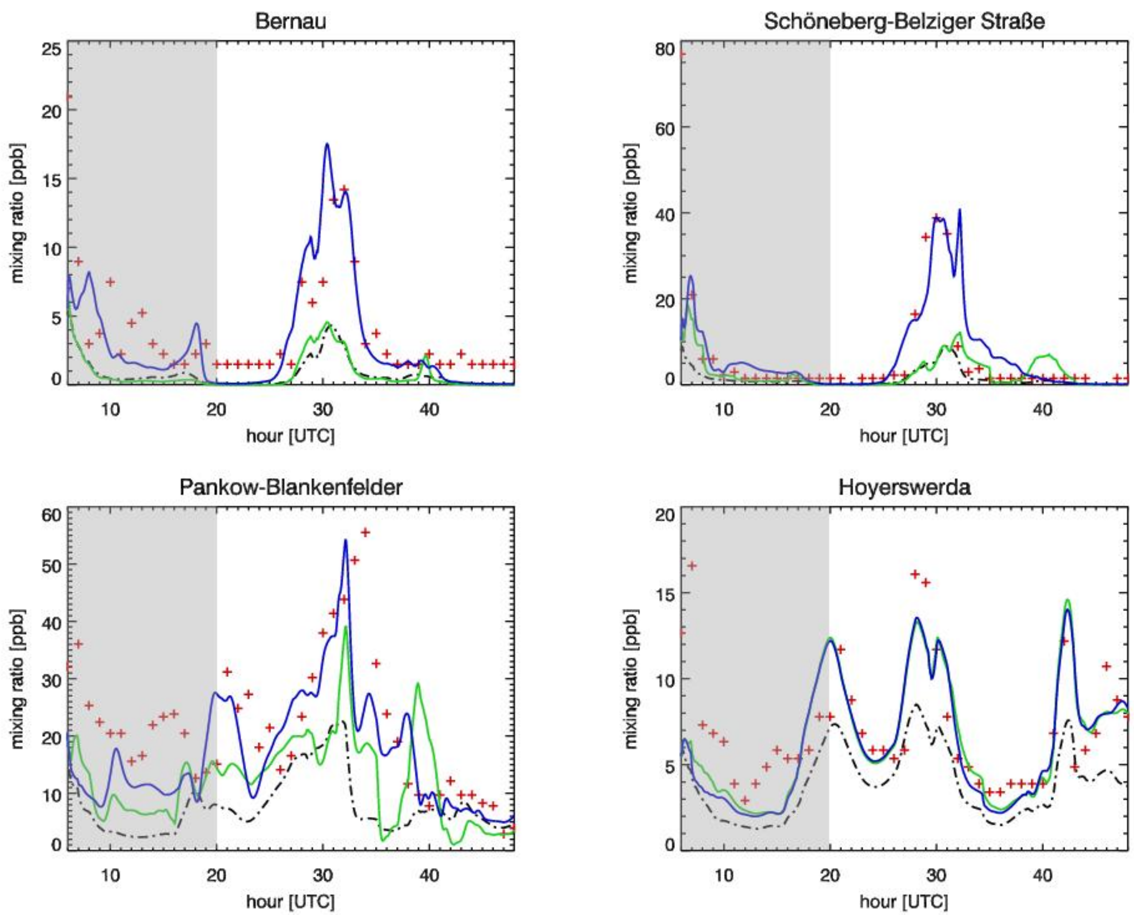
(20.→21. 07.1998)



Some BERLIOZ examples of NOx assimilation (20.→21. 07.1998)

NO

NO_2



Time series for selected NOx stations on nest 2.

+ observations,

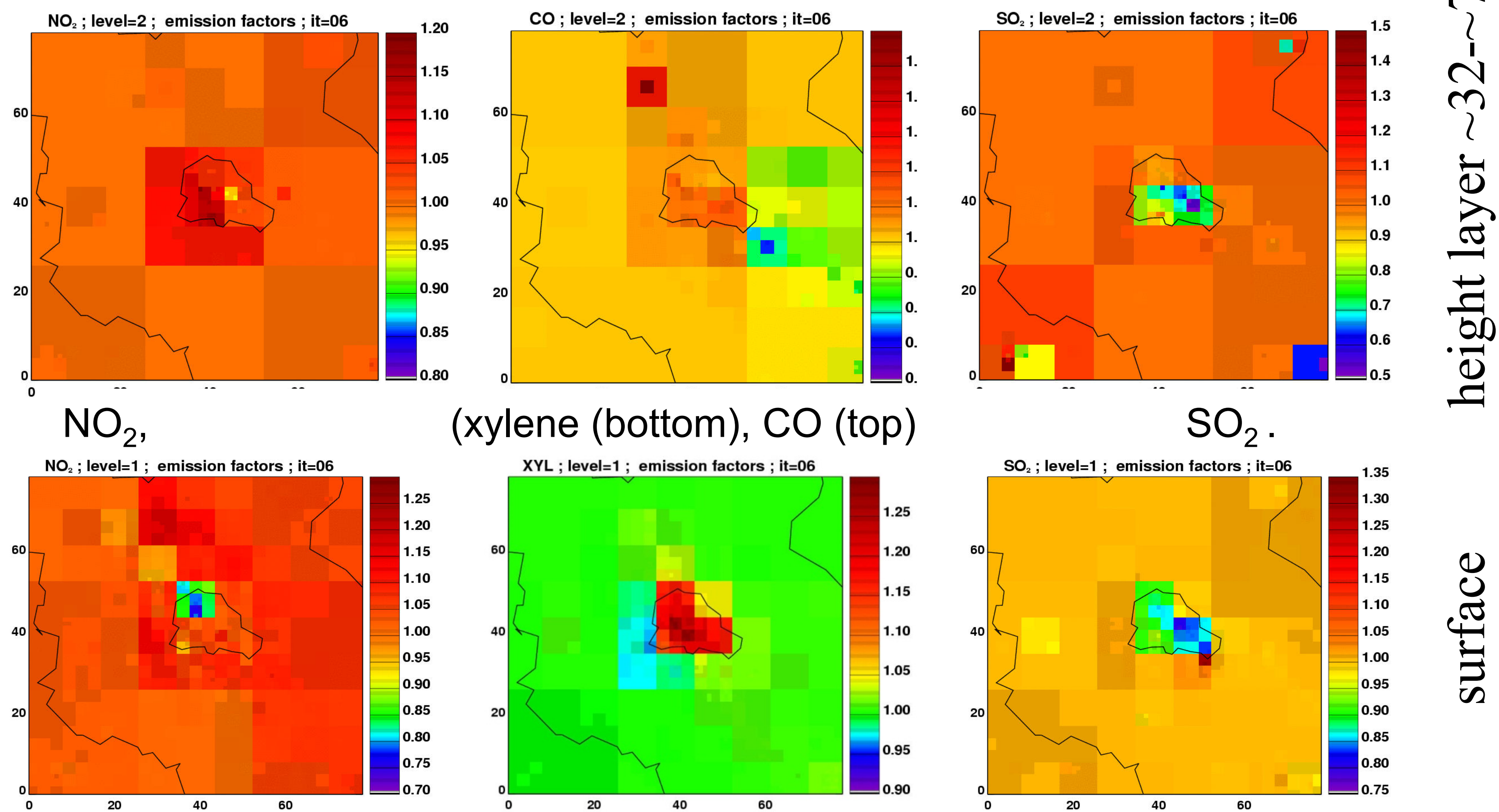
-- - no assimilation,

-_____ N1 assimilation (**18 km**),

-_____N2 assimilation.(**6 km**),

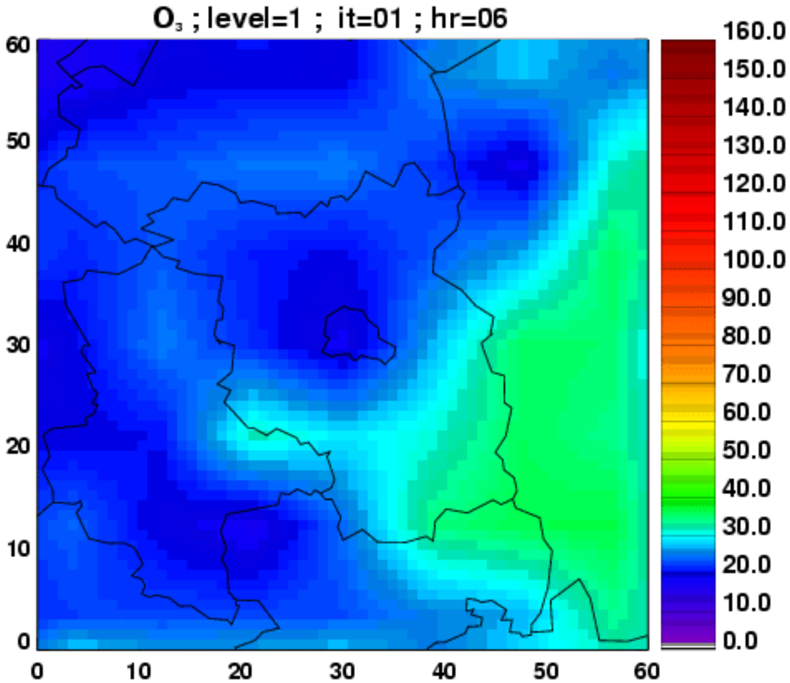
-grey shading: assimilated observations, others forecasted.

Emission source estimates by inverse modelling
Optimised emission factors for Nest 3

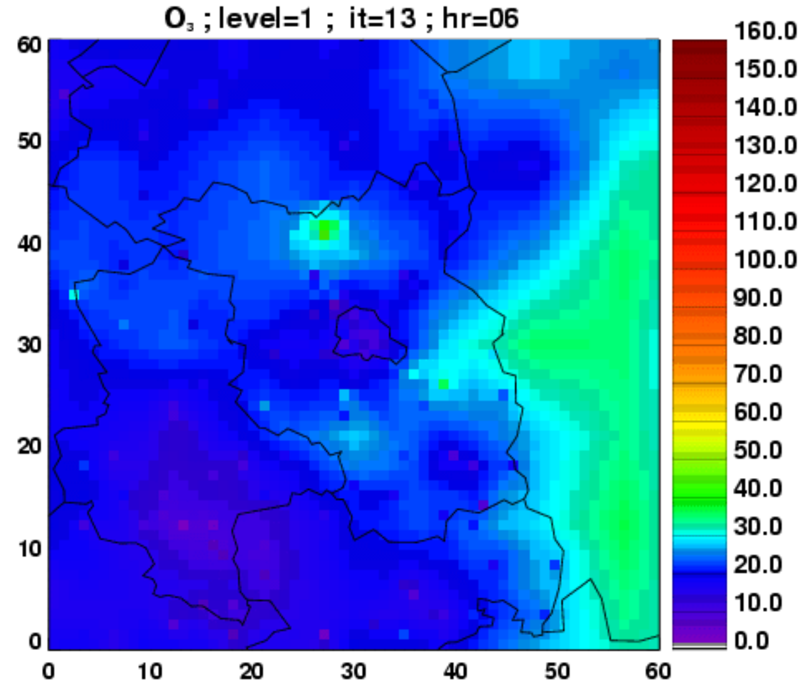


Nest 2: (surface ozone) (20.→21. 07.1998)

without assimilation

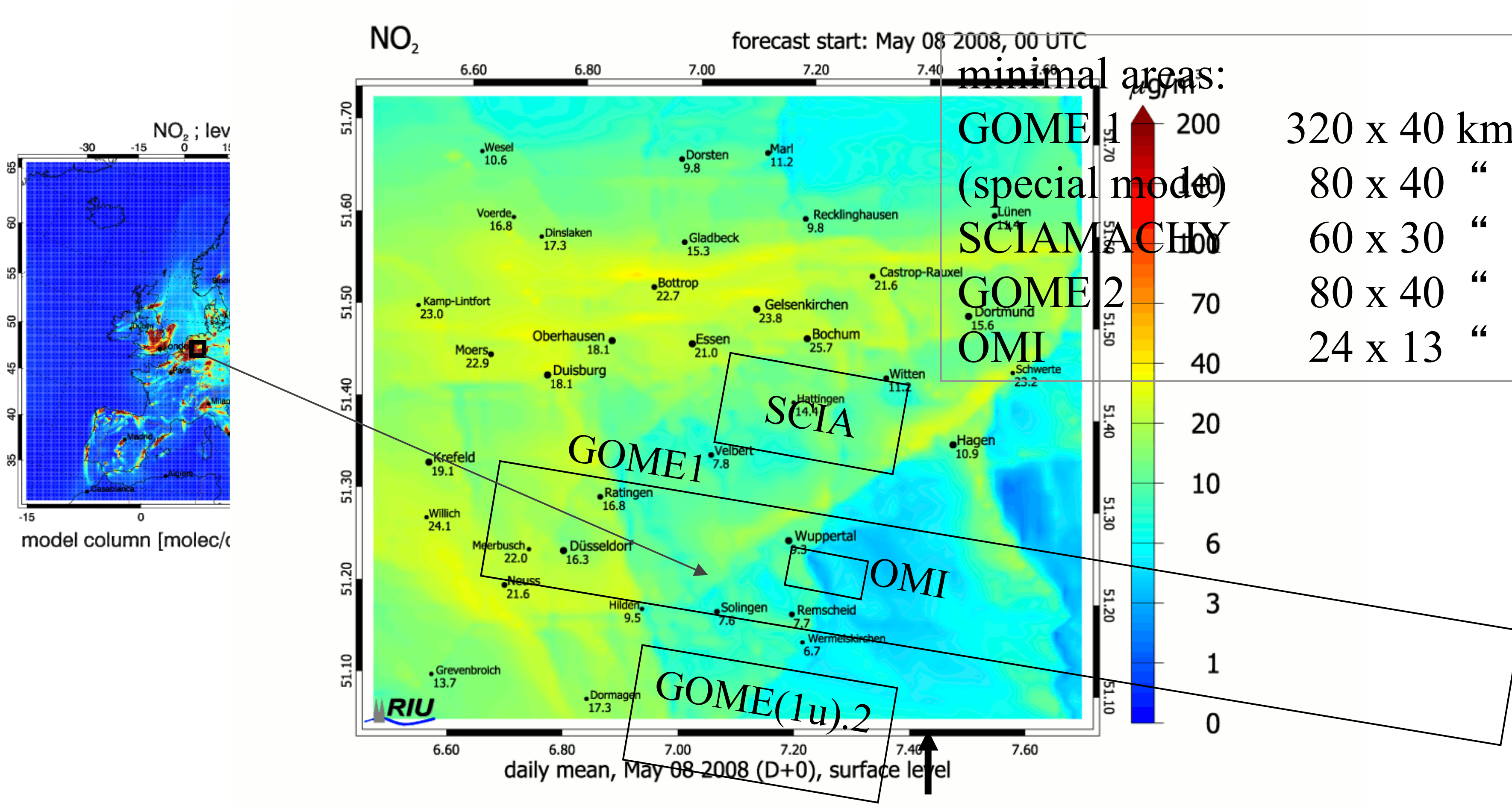


with assimilation



Satellite information:

ESA UV-VIS satellite footprints Ruhr area comparison



minimal areas:

GOME 1	320 x 40 km^2
(special mode)	80 x 40 “
SCIAMACHY	60 x 30 “
GOME 2	80 x 40 “
OMI	24 x 13 “

Ruhr area domain   90 x 80 km^2

(~12 000 000 inhabitants)

Error variances applied for period 1.-10.7.2006 over model domain

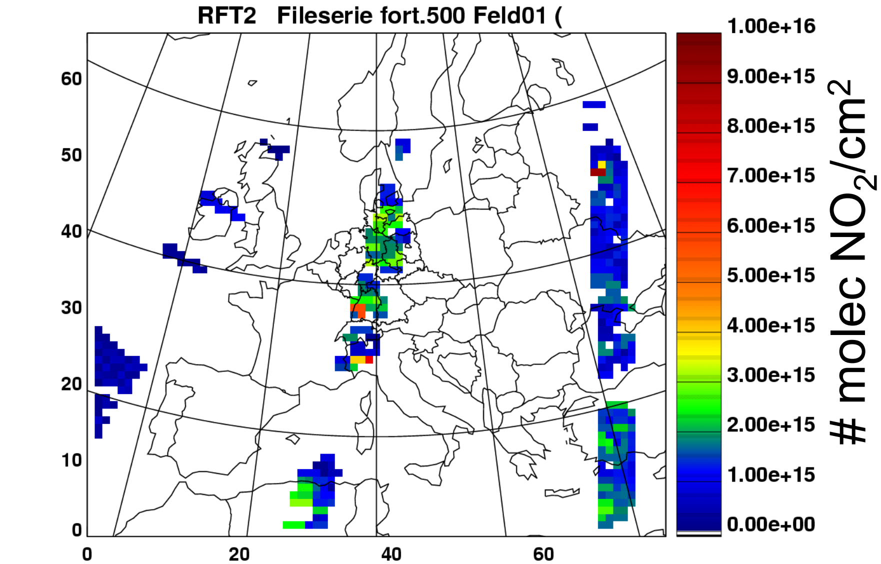
NO2 columns from KNMI data files: **R** (diagonal)

molecules/ cm^2	E(y)	E(Δy)
OMI	$1.4*10^{15}$	$0.8*10^{15}$
SCIAMACHY	$1.2*10^{15}$	$0.9*10^{15}$

Forecast error covariances **B** schematic formula

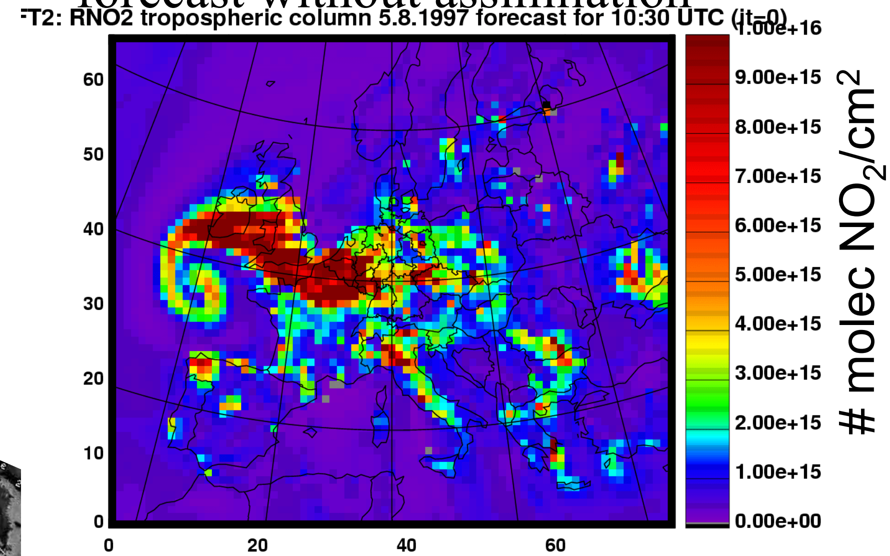
$$\mathbf{B}_{ii}\ (spec,lev) = \max\{1\ ppb, 0.8*var(spec,lev),\ 0.5\ \max(spec,lev)\}$$

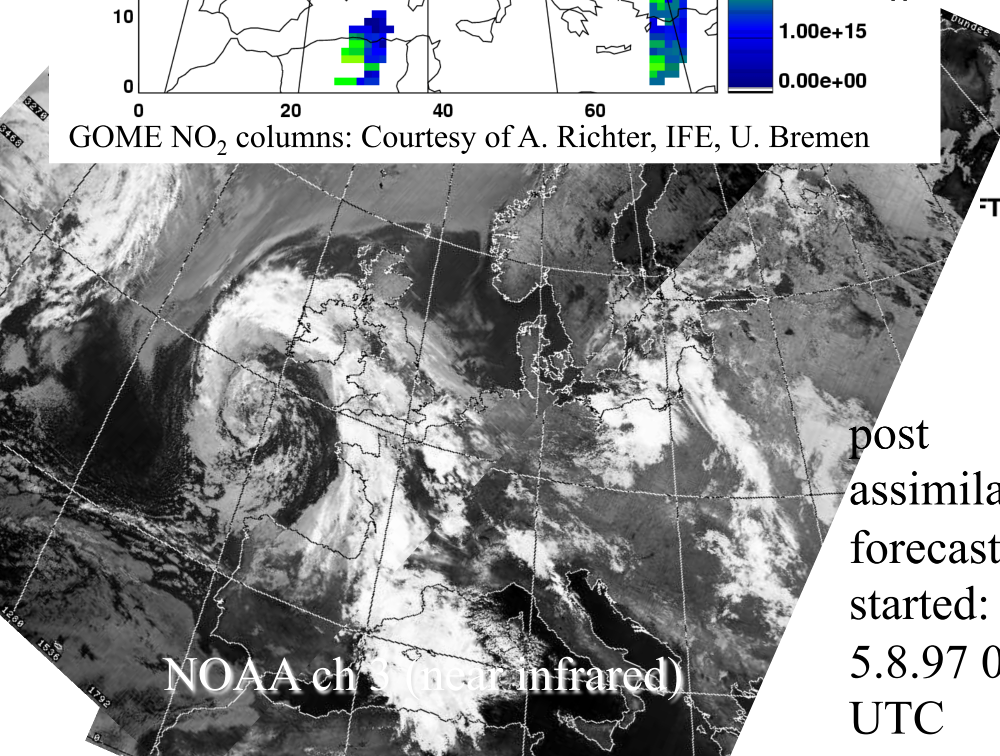
Assimilation of GOME NO_2 tropospheric columns, 5.8.1997



GOME NO_2 columns: Courtesy of A. Richter, IFE, U. Bremen

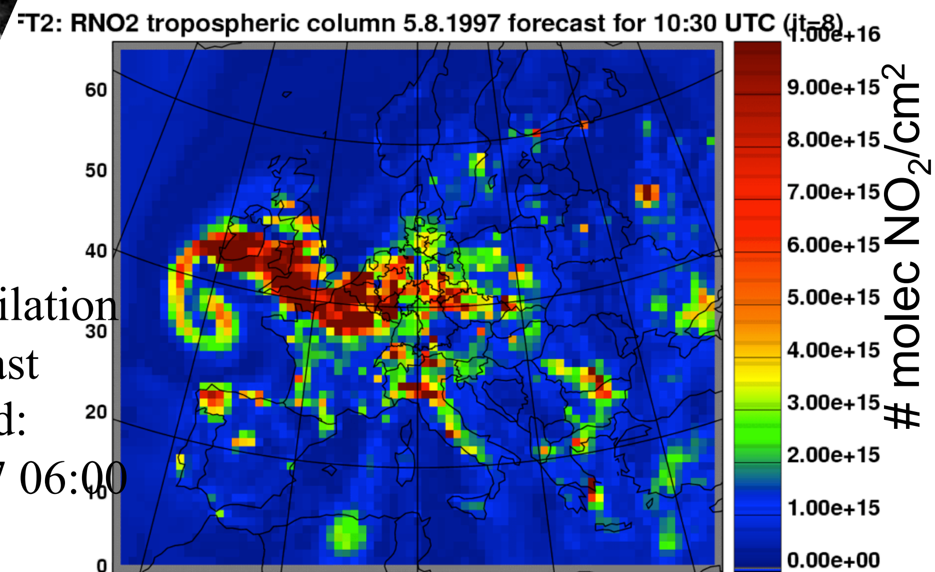
forecast without assimilation





NOAA ch 3 (near infrared)

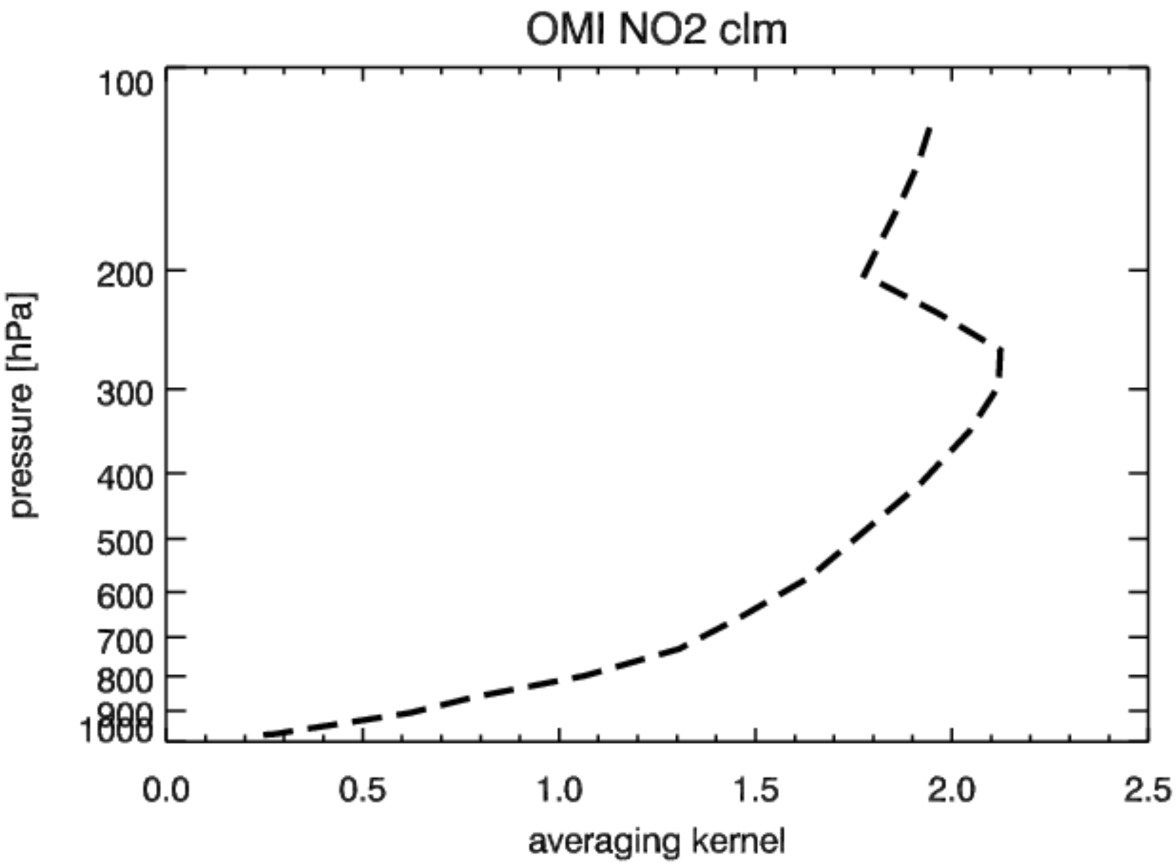
post assimilation forecast started: 5.8.97 06:00 UTC



4. Focus: joint emission rate initial value optimisation

Do NO2 columns profice useful information?

Average OMI averaging kernel profile over model domain for July 9th, 2006



model domain mean averaging kernel.

Exploitation of NO2 column averaging kernel information

- AK profile mostly dependent on optical properties of the atmosphere (cloud cover), rather than NO2

- typical maximal sensitivity **above** the boundary layer

- **does not allow** a clear distinction between PBL or lower free troposphere pollution burden

4. Focus: joint emission rate initial value optimisation

How to proceed to obtain benefit from trop. column integral information?

(A typical problem of Inverse Modelling by Integral Equations)

Two more specific questions:

- When is it justified to project averaging kernel information to the surface?

- Can this be done without heuristics, destroying the BLUE property of the assimilation algorithm?

Observation operator H

Formally an integral equation to be solved for vertical NO_2 molecule density function x (σ vertical coordinate)

$$y = \int_1^0 w(\sigma)x(\sigma)d\sigma$$

Discretisation

$$y = \sum^K h_k x_k$$

At the minimum $\mathbf{x} =: \mathbf{x}_a$

$$d\mathbf{x}_a := \mathbf{x}_a - \mathbf{x}_b = (\mathbf{B}_0^{-1} + \mathbf{H}^T\mathbf{R}^{-1}\mathbf{H})^{-1}\mathbf{H}^T\mathbf{R}^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x}_b]\right\}$$

$$= \mathbf{B}\mathbf{H}^T(\mathbf{R} + \mathbf{H}\mathbf{B}\mathbf{H}^T)^{-1}\left\{\mathbf{y}^0 - H[\mathbf{x}_b]\right\}$$

For scalar column retrieval:

$$d\mathbf{x}_a^c = \underbrace{\mathbf{B}\mathbf{h}^T(r+b)^{-1}}\left\{y^0 - H[\mathbf{x}_b]\right\}$$

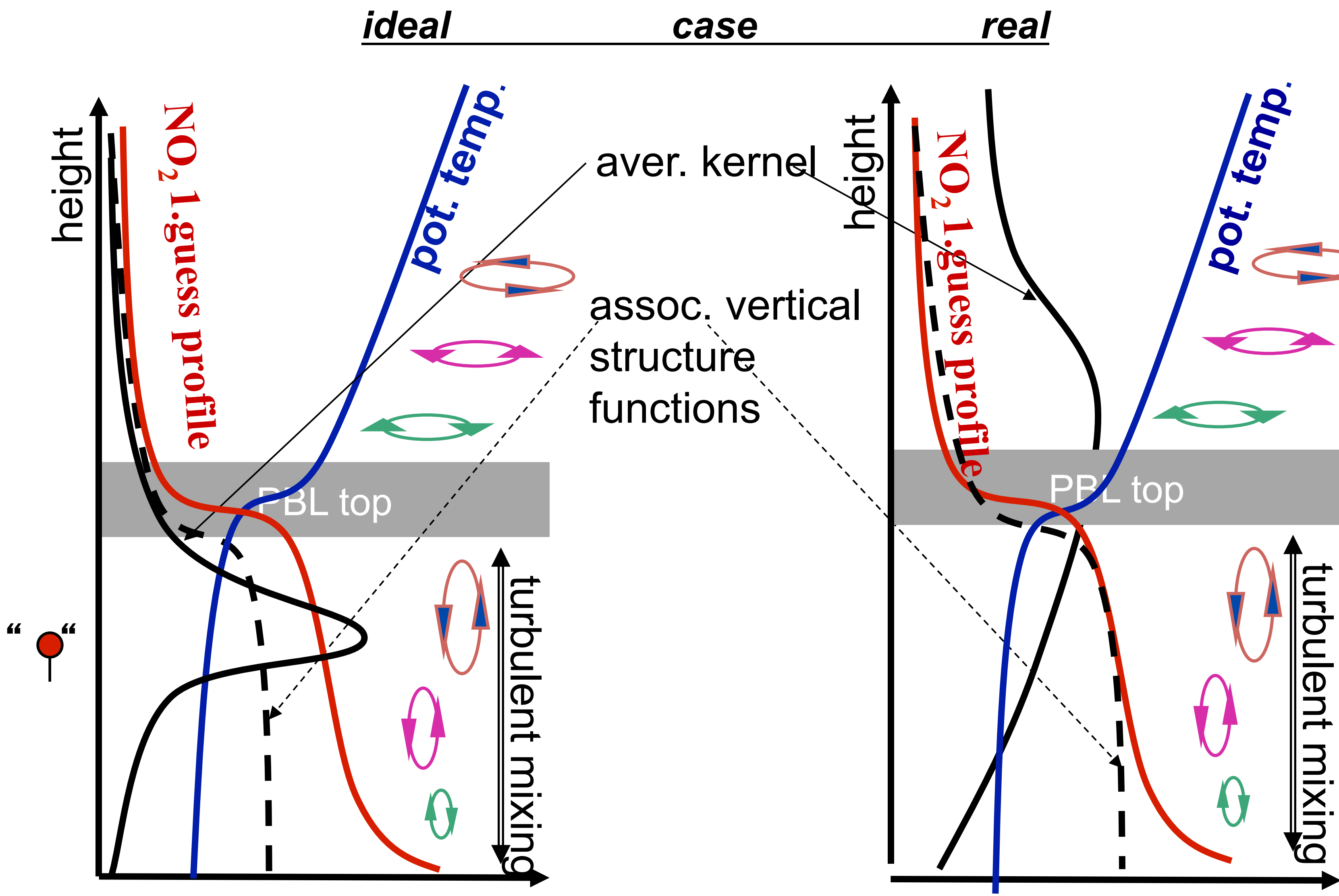
adjoint representer

→ vertical structure function in B essential!

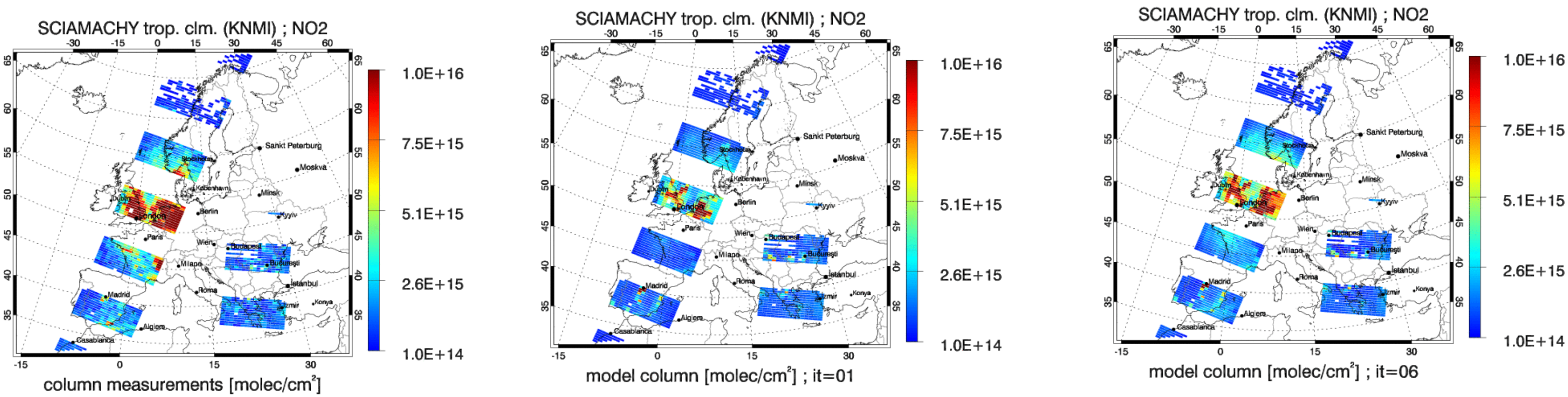
4. Focus: joint emission rate initial value optimisation

Vertical structure function:

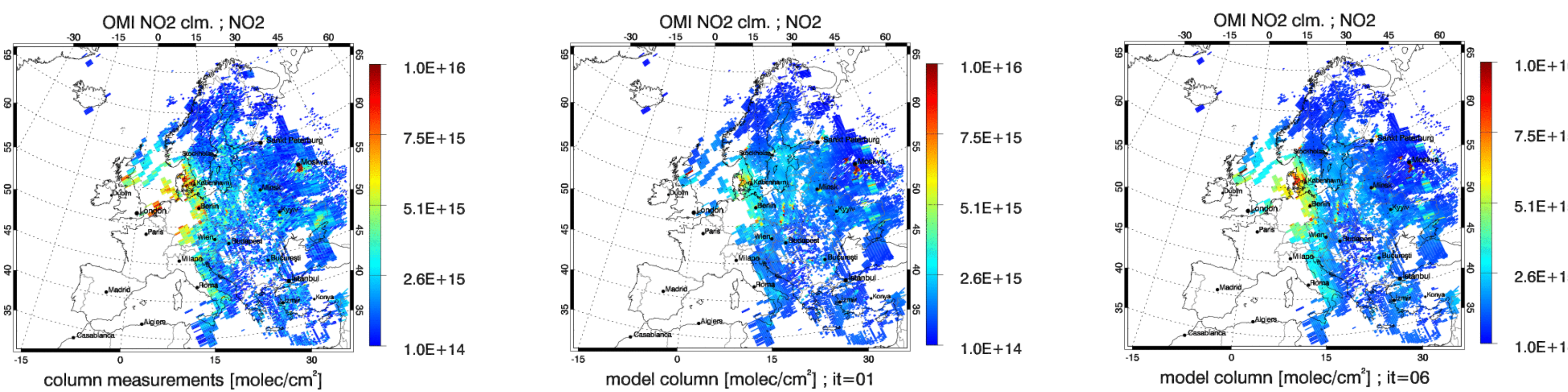
Extending the information from observation location by vertical exchange of polutants and information



Comparison of NO2 tropospheric columns in molecules/cm2 for July 6th, 2006, 09-12 UTC.

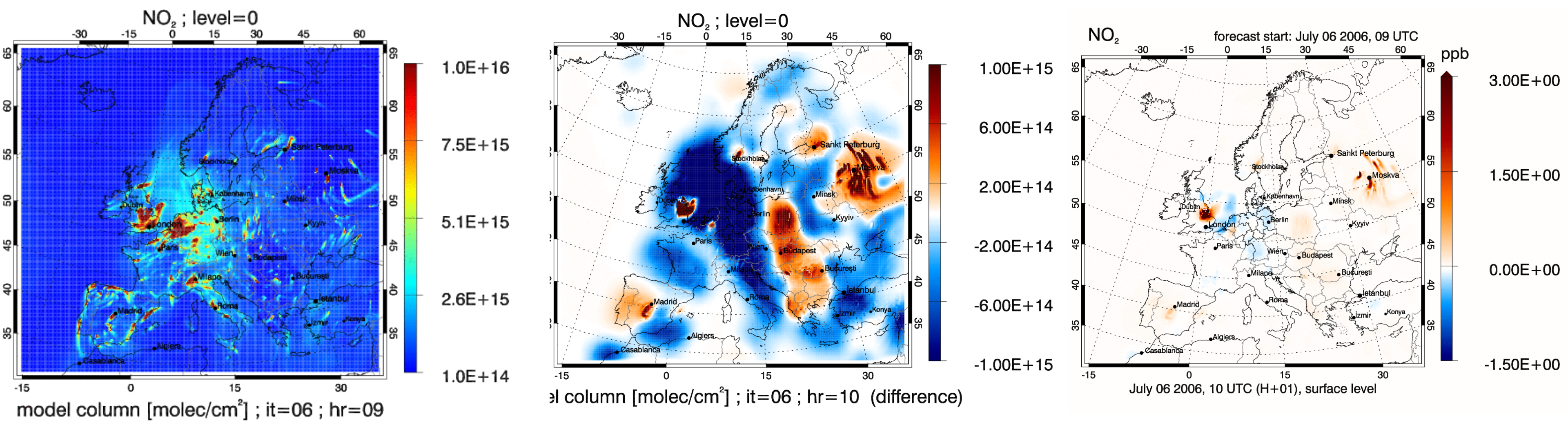


assimilated values (**y**); :EURAD forecasted ($\mathbf{Hx}_b$); column analyses ($\mathbf{Hx}_a$).



4. Focus: joint emission rate initial value optimisation

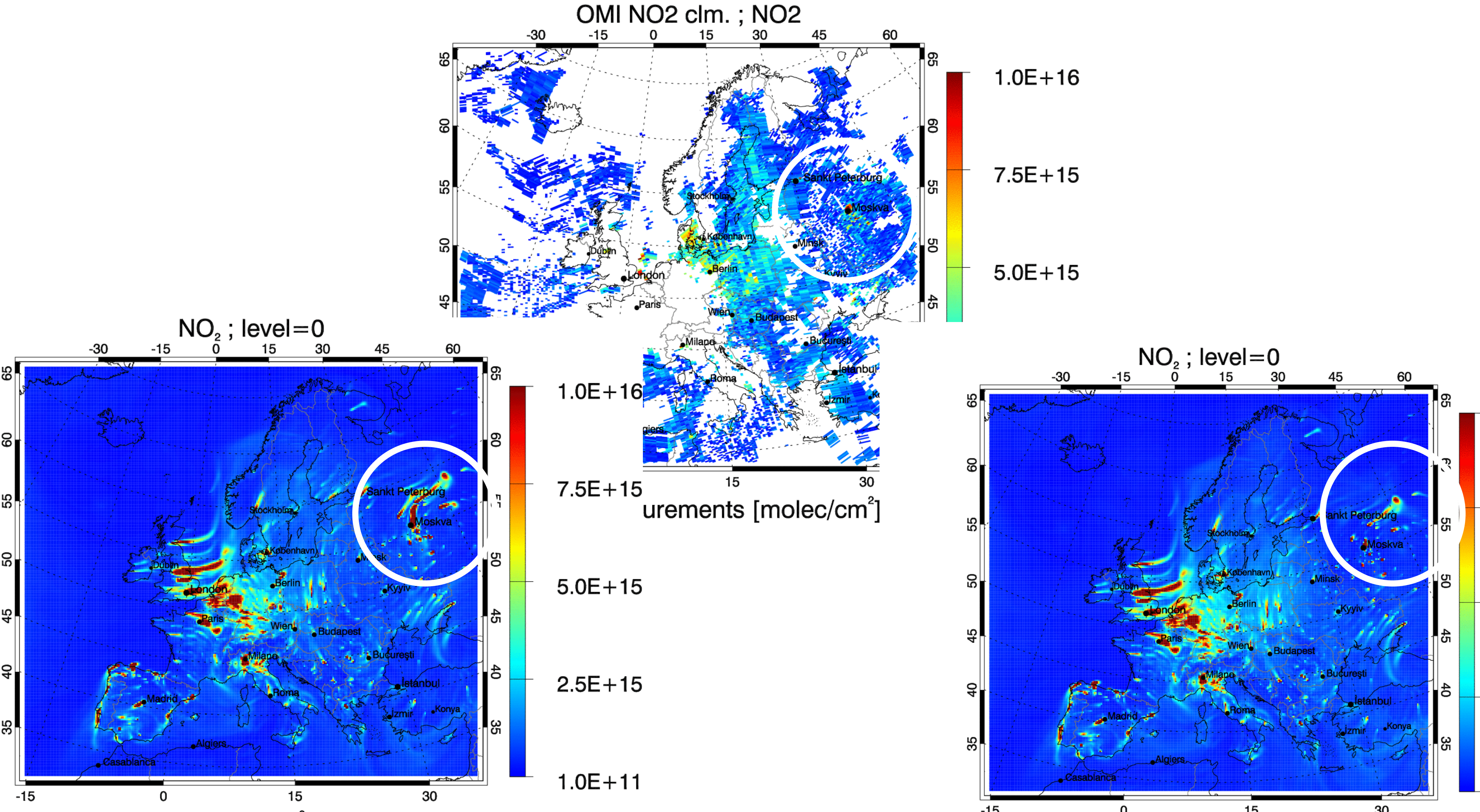
Data assimilation result in terms of tropospheric columns for July 6th, 2006. NO2 model columns based on OMI and SCIAMACHY assimilation within interval, 09-12 UTC.



Difference field giving implied changes for tropospheric columns by assimilation (middle), and induced surface concentration changes by NO2 ppb (right)

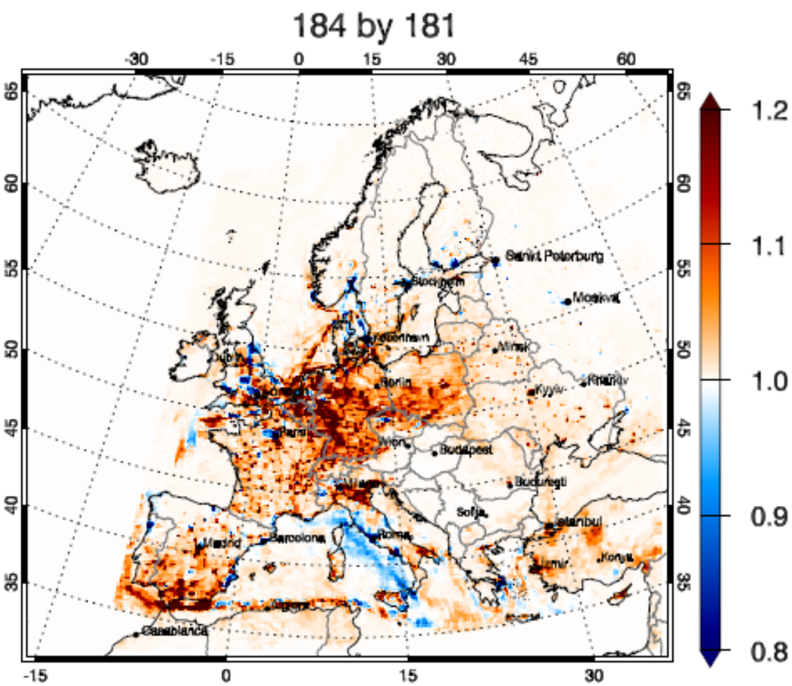
4. Focus: joint emission rate initial value optimisation

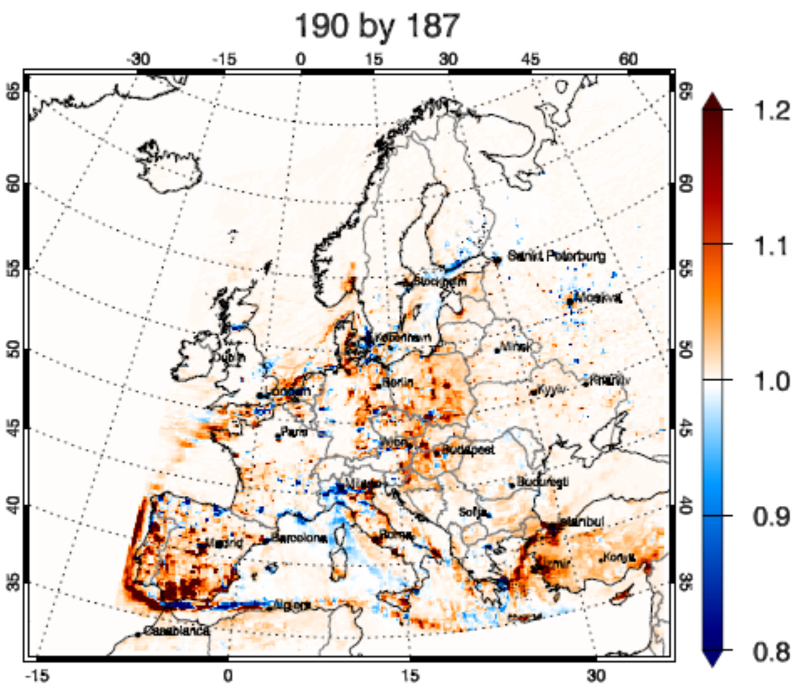
Data assimilation result in terms of tropospheric columns for **July 7th**, 2006. NO2 model columns based on OMI and SCIAMACHY assimilation within the assimilation interval, 09-12 UTC.



Emission rate optimisation factors for NO2 after assimilation of

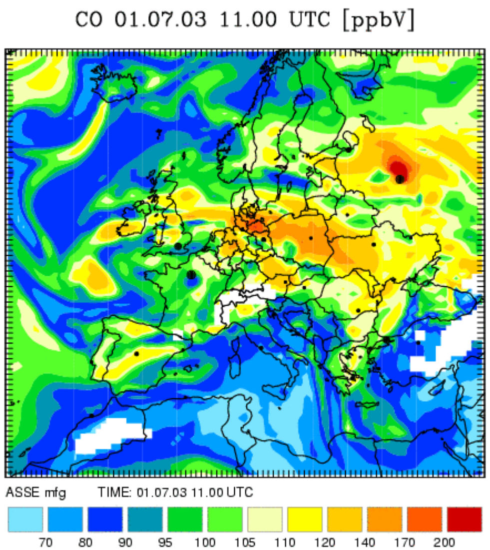
OMI retrieved NO2 tropospheric columns

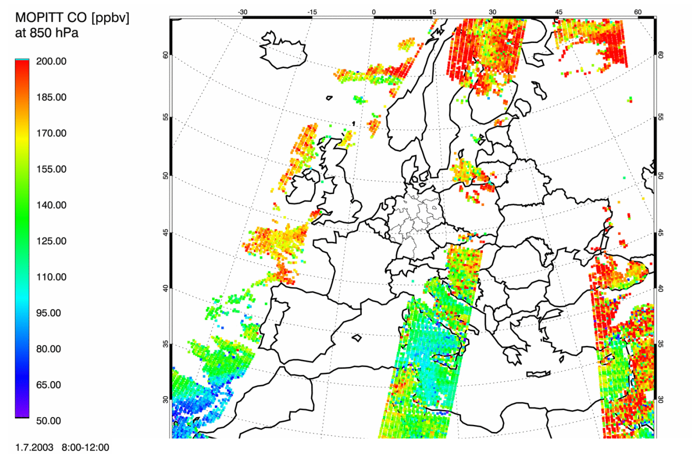




2 x 4 days assimilation sequence. Left panel shows results after assimilation procedures from July 1.-4. 2006, right panel for July 7.-11., 2006. OMI data from KNMI

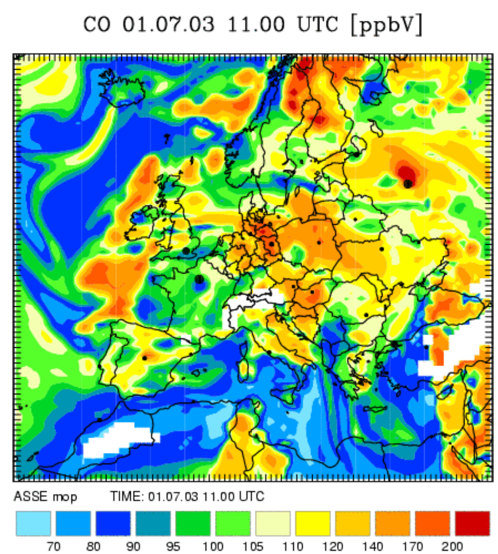
MOPITT tropospheric CO assimilation





retrieved 850 hPa concentrations in ppbV.

first guess at 11:00 UTC;.



4D-Var analysis for the same time;

850 hPa, 1 July. 2003

Note: Colour codes are not fully consistent!

Contents

1. Introductory remarks
2. **Where is atmospheric chemistry a challenge for data assimilation ? (focal topics)**
    1. strongly anisotropic covariances
    2. diversity of chemical regimes: validation
    3. **varying value of observations**
    4. **more than initial values to be optimised**
3. **Aerosol data assimilation**

Example:
Aerosol Chemistry in
MADE

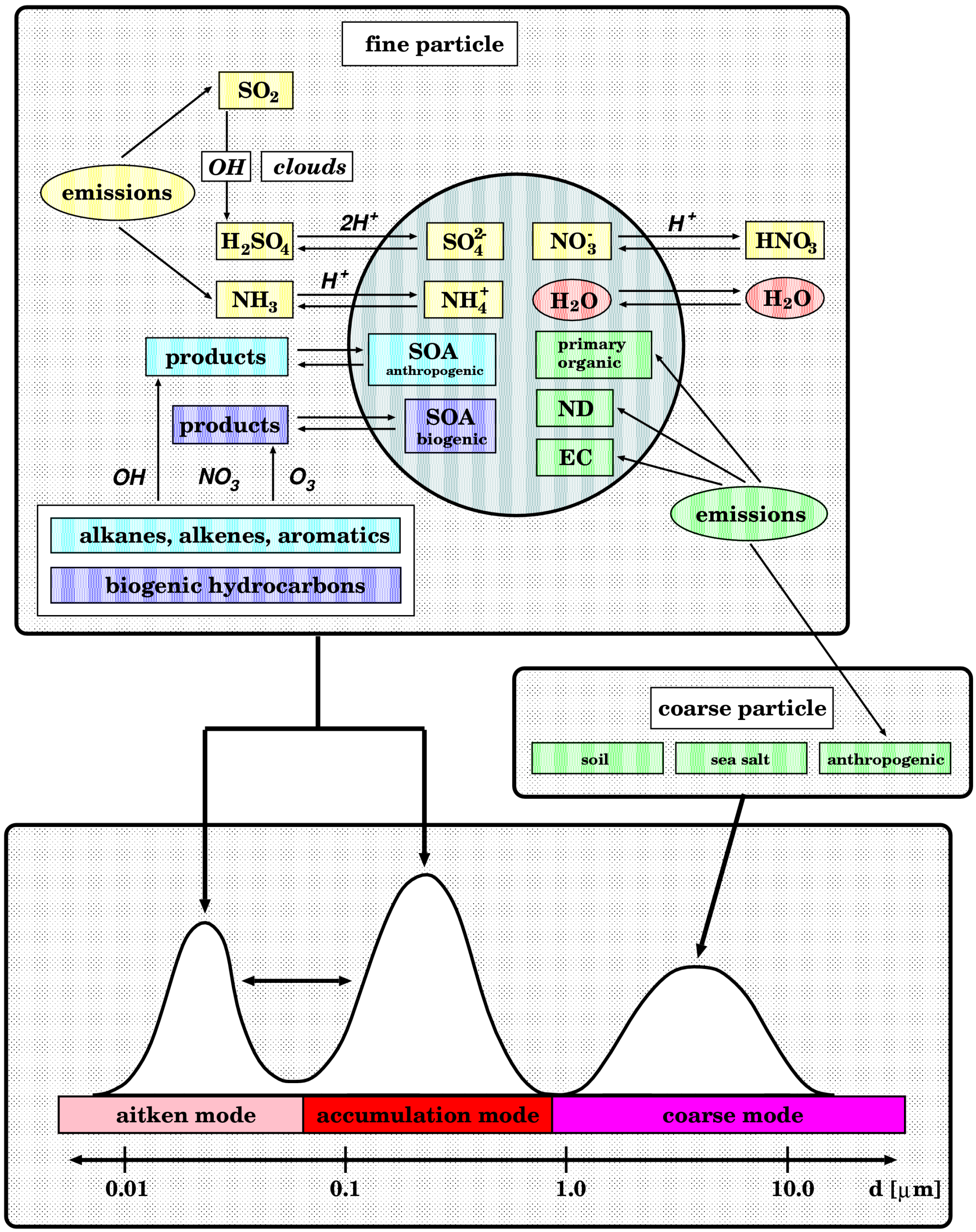
Modal **A**erosol **D**ynamics for
EURAD/**E**urope
(Ackerman et al., 1998, Schell 2000)

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

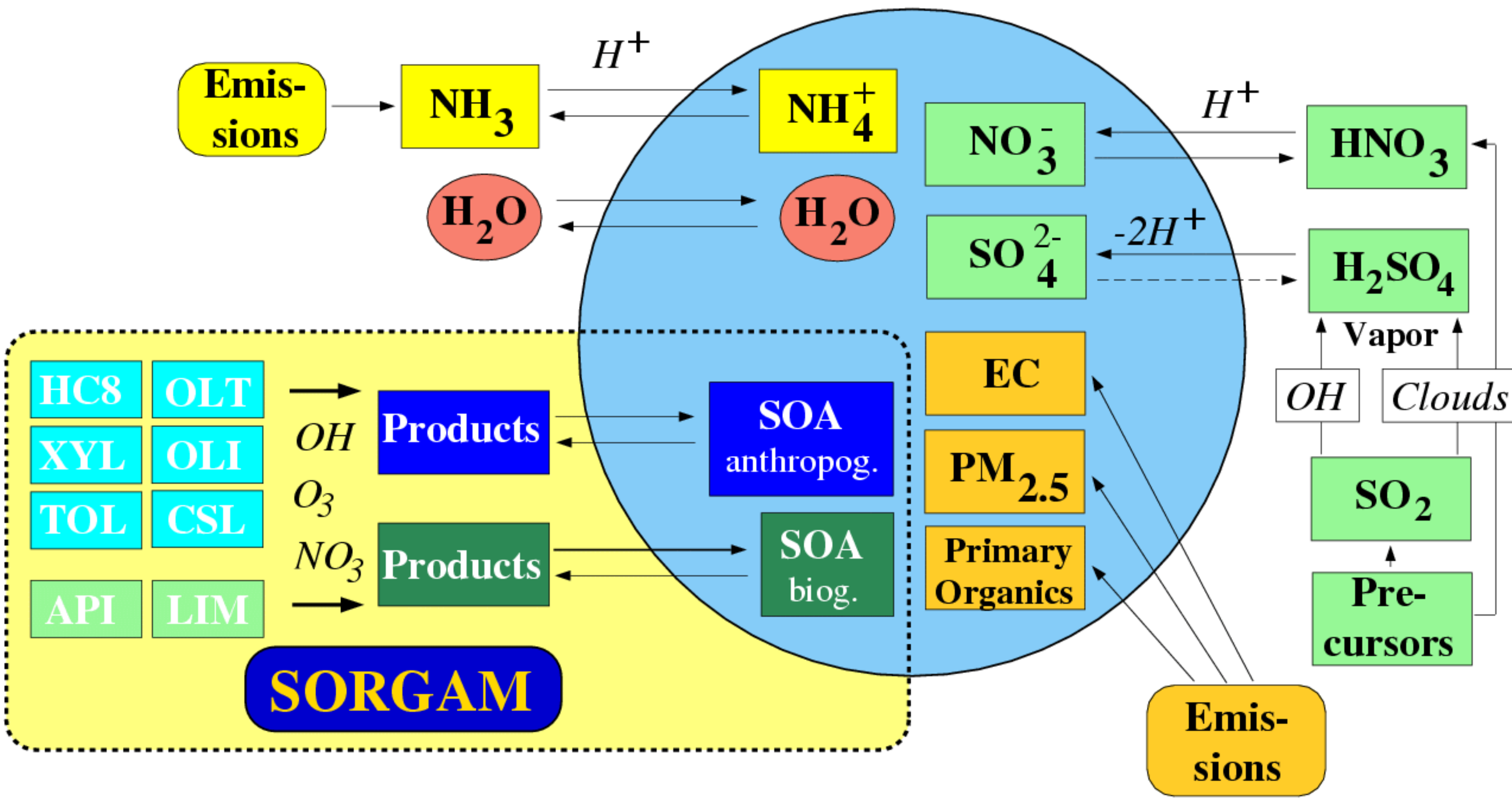
M_i^k:= k^{th} Moment of i^{th} Mode

assimilation of aerosol
By satelite retrievals: e.g.
Bridge from optical to chemical properties

MERIS MODIS
AATSR+SCIAMACHY,...



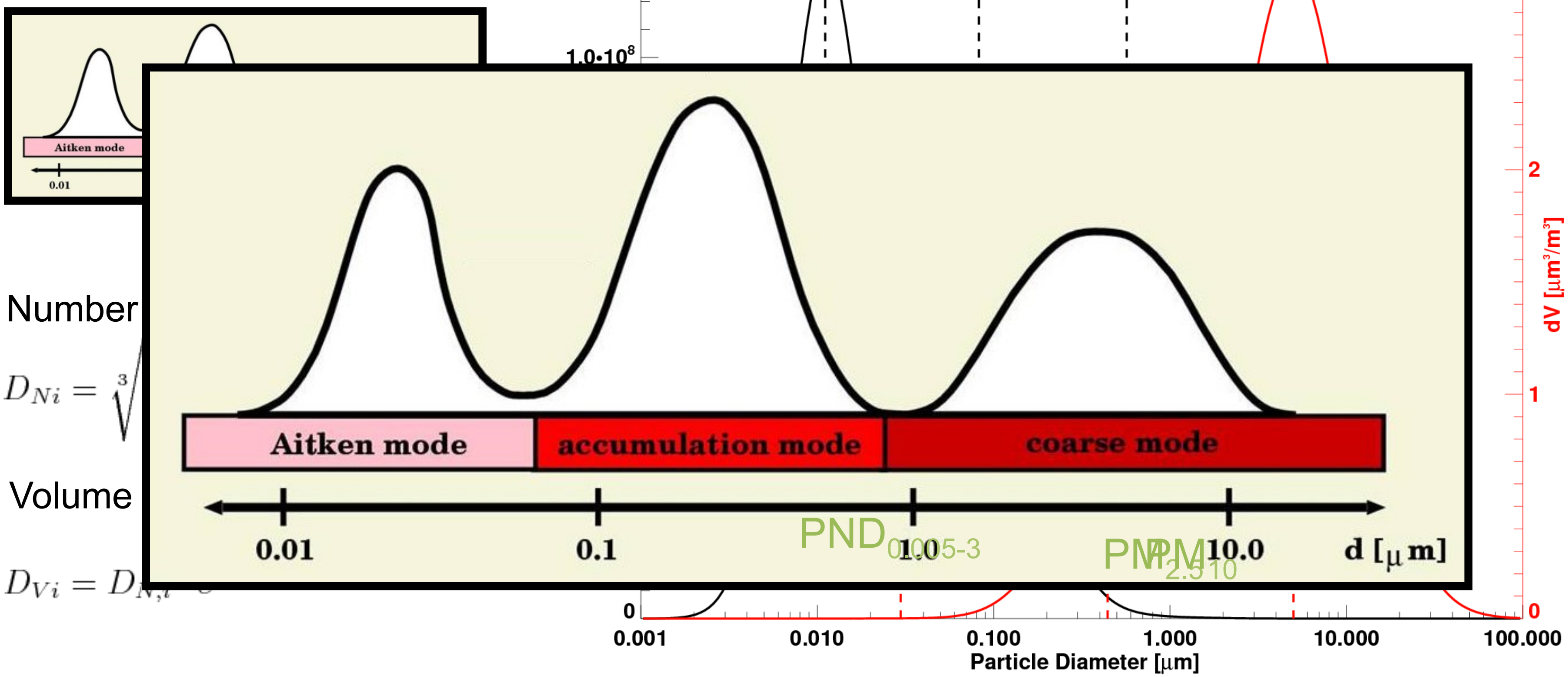
Secondary ORGanic Aerosol Model (SORGAM)
Biogenic and anthropogenichydrocarbons to particles



Modal representation of particles

Particulate Matter (PM_x) and Particle Number Density ($PND_{0.005-3.0}$)

Trimodal Log-normal distribution



Mass/Number Concentrations

Integrator →

← Adjoint Integrator

PM_x / PND

Assimilation of Aerosol observations

- In situ:

  EEA Airbase: Database of groundstations of EU member countries & states:
  - 450 stations for PM_{10} (2003)
  - No $PM_{2.5}$. (4 stations in UK only)
- Satellite measurements:

  SYNAER (SYNergetic AErosol Retrieval, DLR-DFD, [Holzer-Popp, 2001])*
  - combines GOME&ATSR-2, SCIAMACHY&AATSR measurements aboard ERS-2/ENVISAT
  - ATSR-2/AATSR:
    dark field detection, BLAOT (Boundary Layer Aerosol Optical Thickness) and albedo are calculated
  - GOME/SCIAMACHY:
    Provides $PM_{0.5}$, $PM_{2.5}$ and PM_{10} columns and its composition (6 intrinsic species)

SYNAER retrieval algorithm

SYNergetic AErosol Retrieval (Holzer-Popp, 2001)

SYNAER makes combined use of two instruments (Radiometer and Spectrometer) on ERS-2, Envisat, and MetOp:

- ATSR-2/AATSR/AVHRR

  Dark field detection, BLAOT (Boundary Layer AOT), Albedo

- GOME/Sciamachy/GOME-2

  Measured spectra taken to identify aerosol mixture of highest

  probability out of 40 predefined mixtures via a Least Squares

  Method

SYNAER retrieval algorithm

Species Mapping

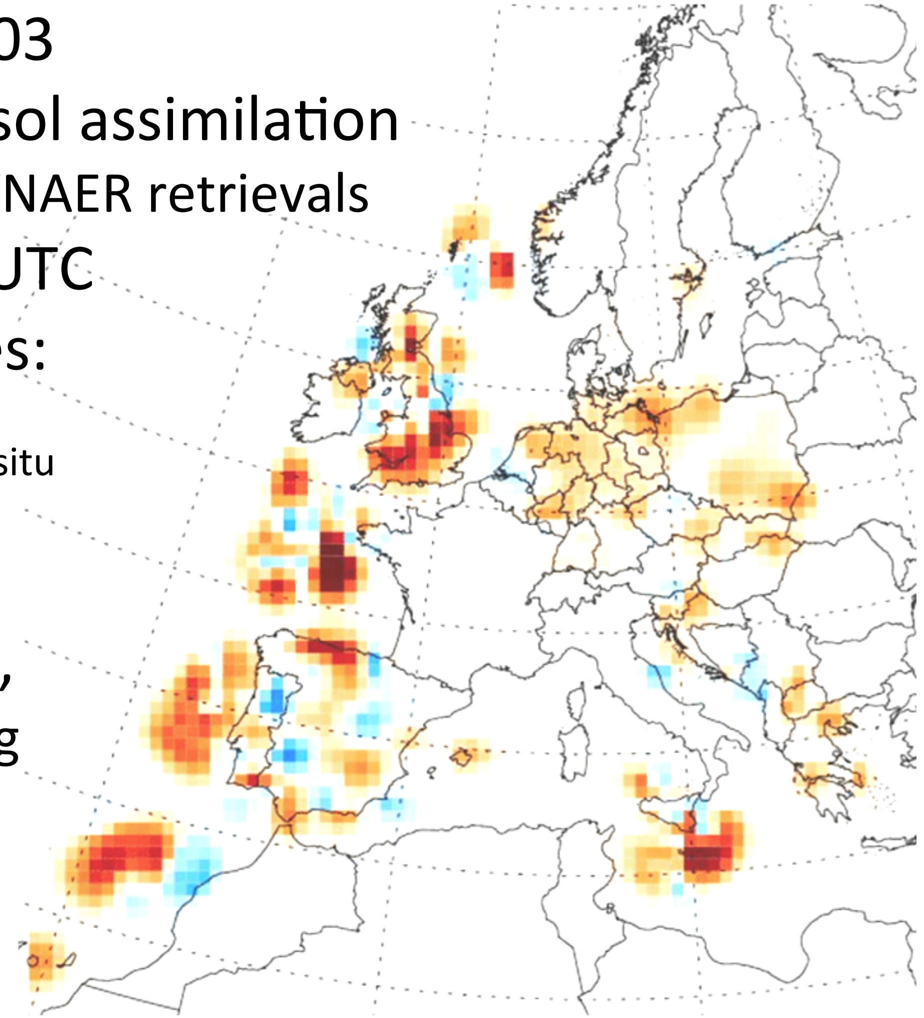
EURAD-IM [µg/m^3]		SYNAER - AOT
SO_4, NH_3, NO_3, H_2O, SOA		WASO (WAter SOluble)
Unidentified PM		INSO (water INSOluble)
Elemental Carbon		SOOT
Sea Salt		SEAS
Mineral Dust		DUST

radiative transfer model →

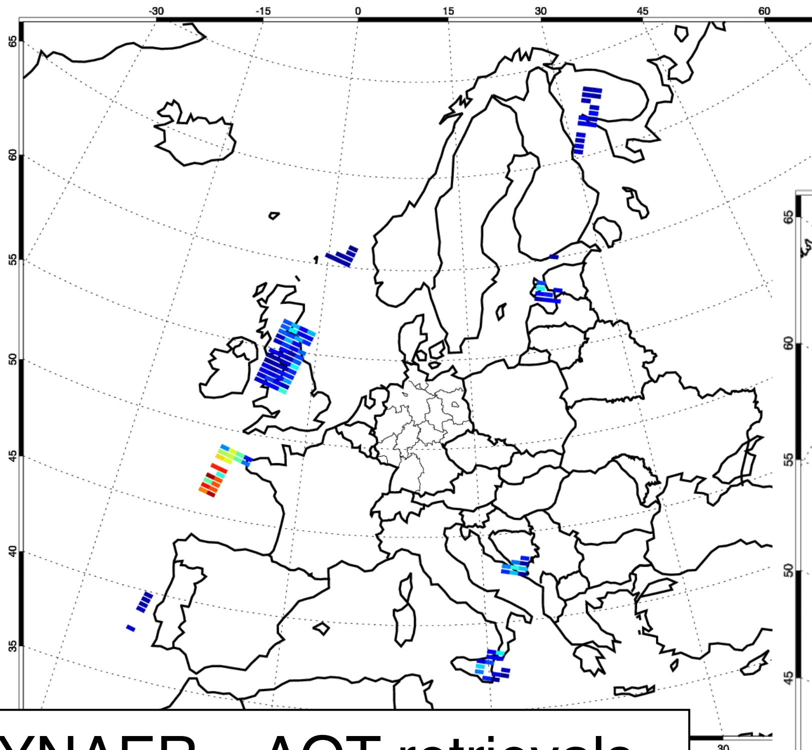
← adjoint radiative transfer model

Case study I: Summer 2003

- Episode   July 1 - July 14, 2003
- Purpose   Validation of aerosol assimilation
  scheme & SYNAER retrievals
- Assimilation   8 – 16(14) UTC
- Data assimilated in 3 sub-cases:
  1. SYNAER-AOT only
  2. SYNAER-AOT & EEA in-situ
  3. EEA in-situ only
- Grid   107 x 97 x 23 Cells,
  45km spacing
- Timestep   600 s



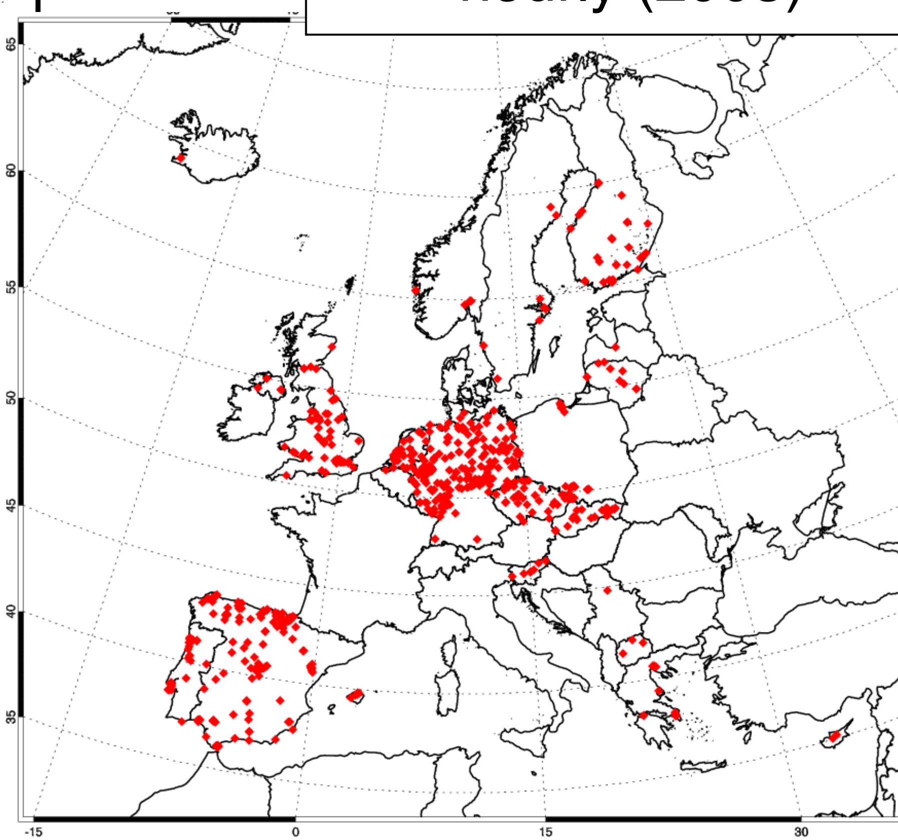
Model Domain and Observations



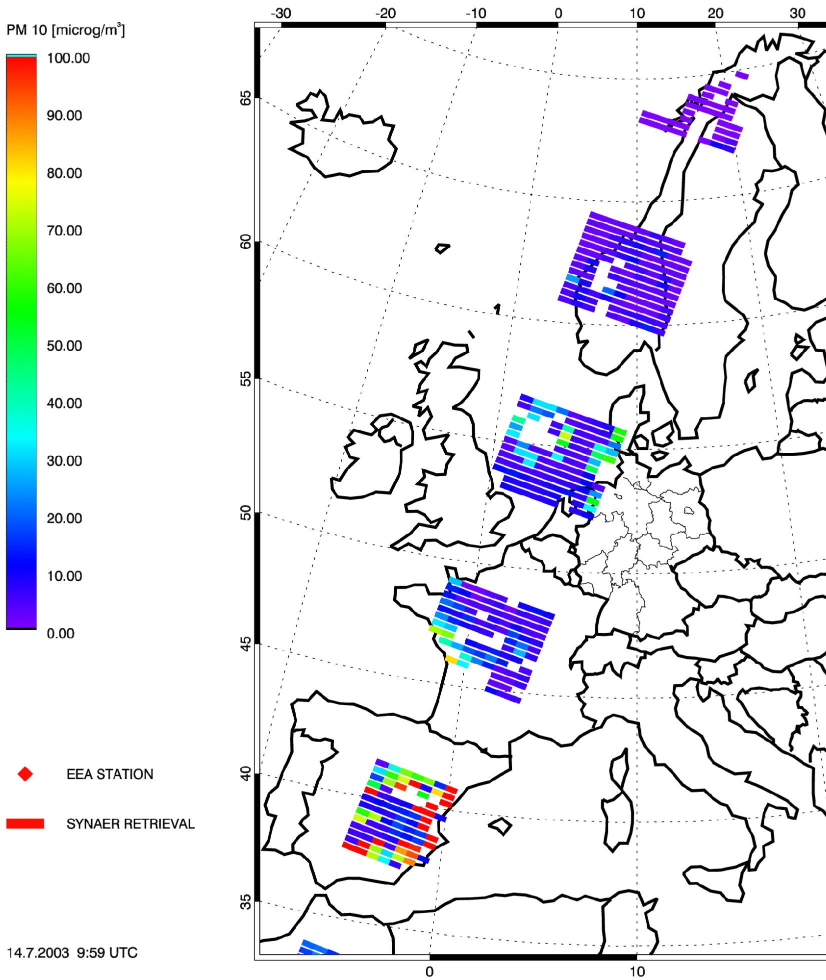
SYNAER – AOT retrievals
2 – 3 traces per day

13 July 2003

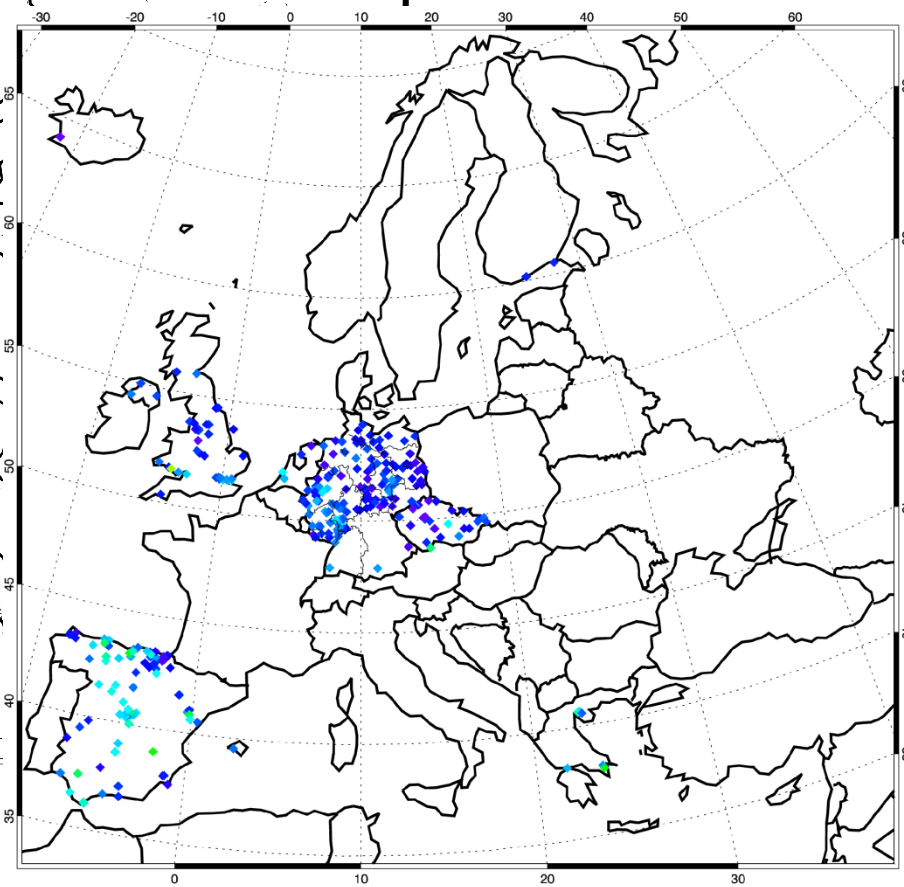
EEA in-situ PM_{10}
hourly (2003)



Aerosol observations (14.7.2003, ~10:00 UTC)

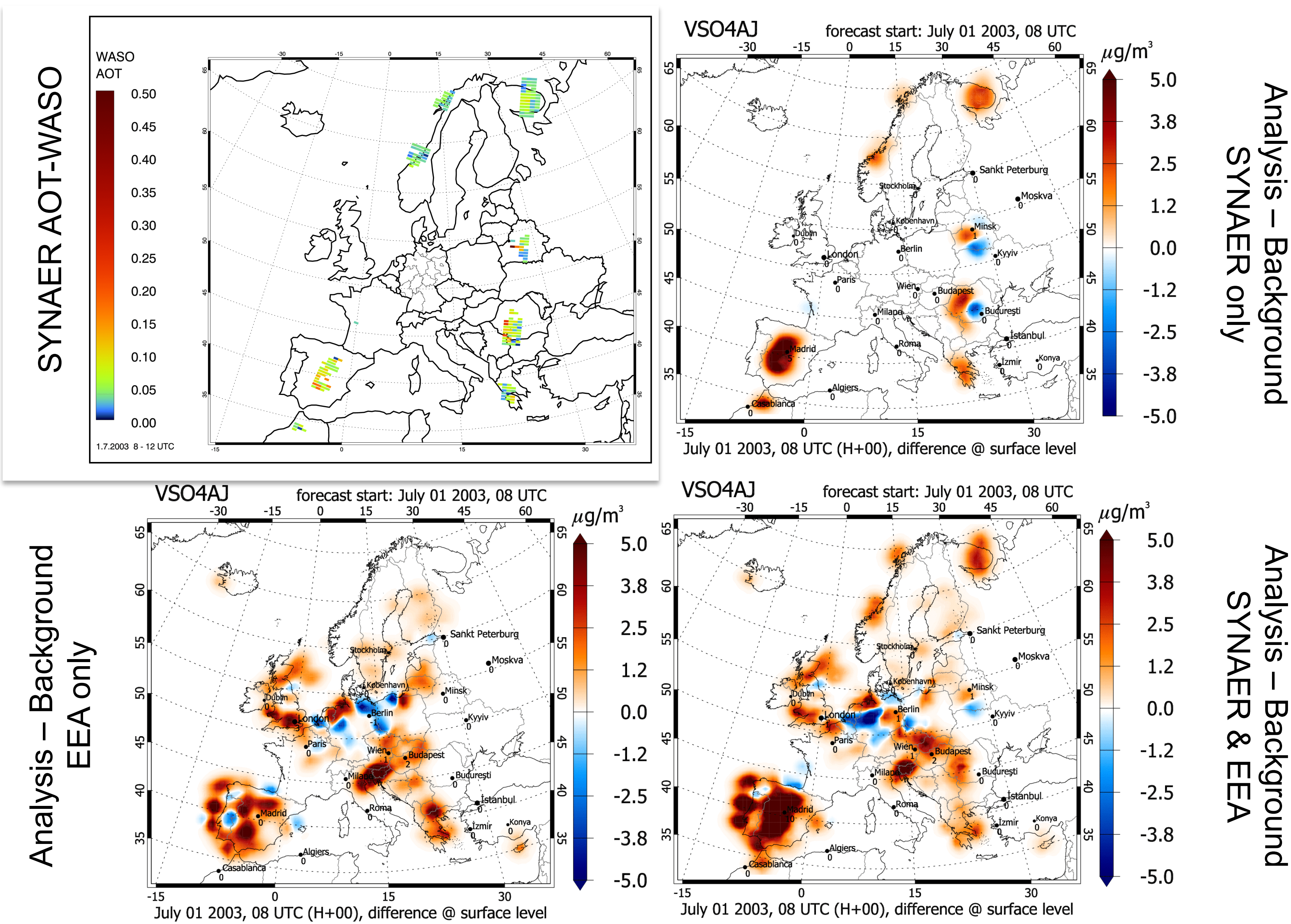


SYNAER PM_{10} retrievals



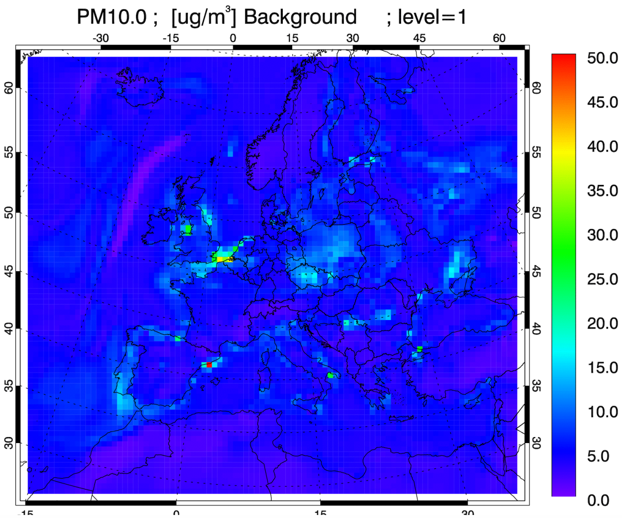
EEA PM_{10} stations

Analysis and increments for acc. SO_4^{2-} July 1, 8 UTC

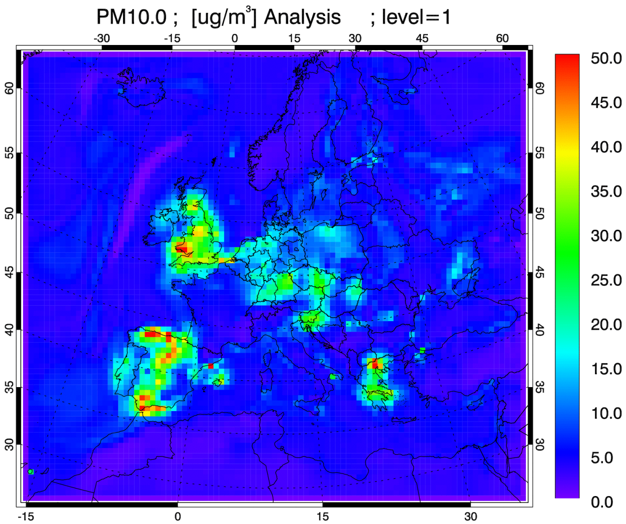


3Dvar aerosol assimilation (13.7.2003)

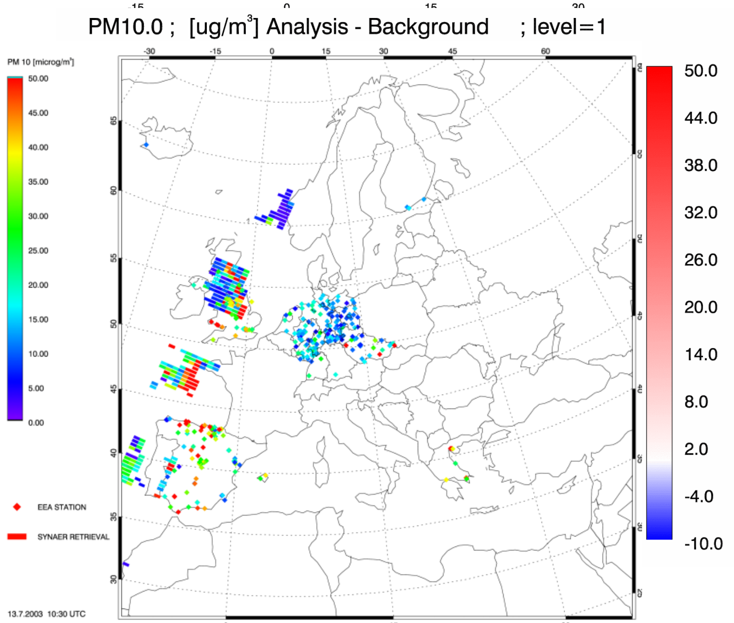
background



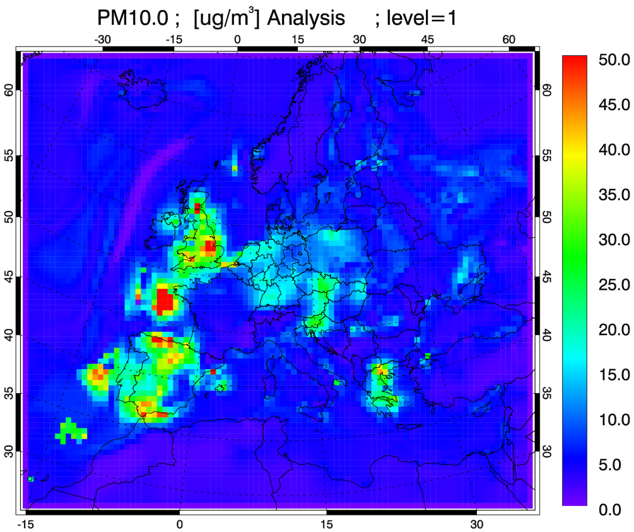
in situ only



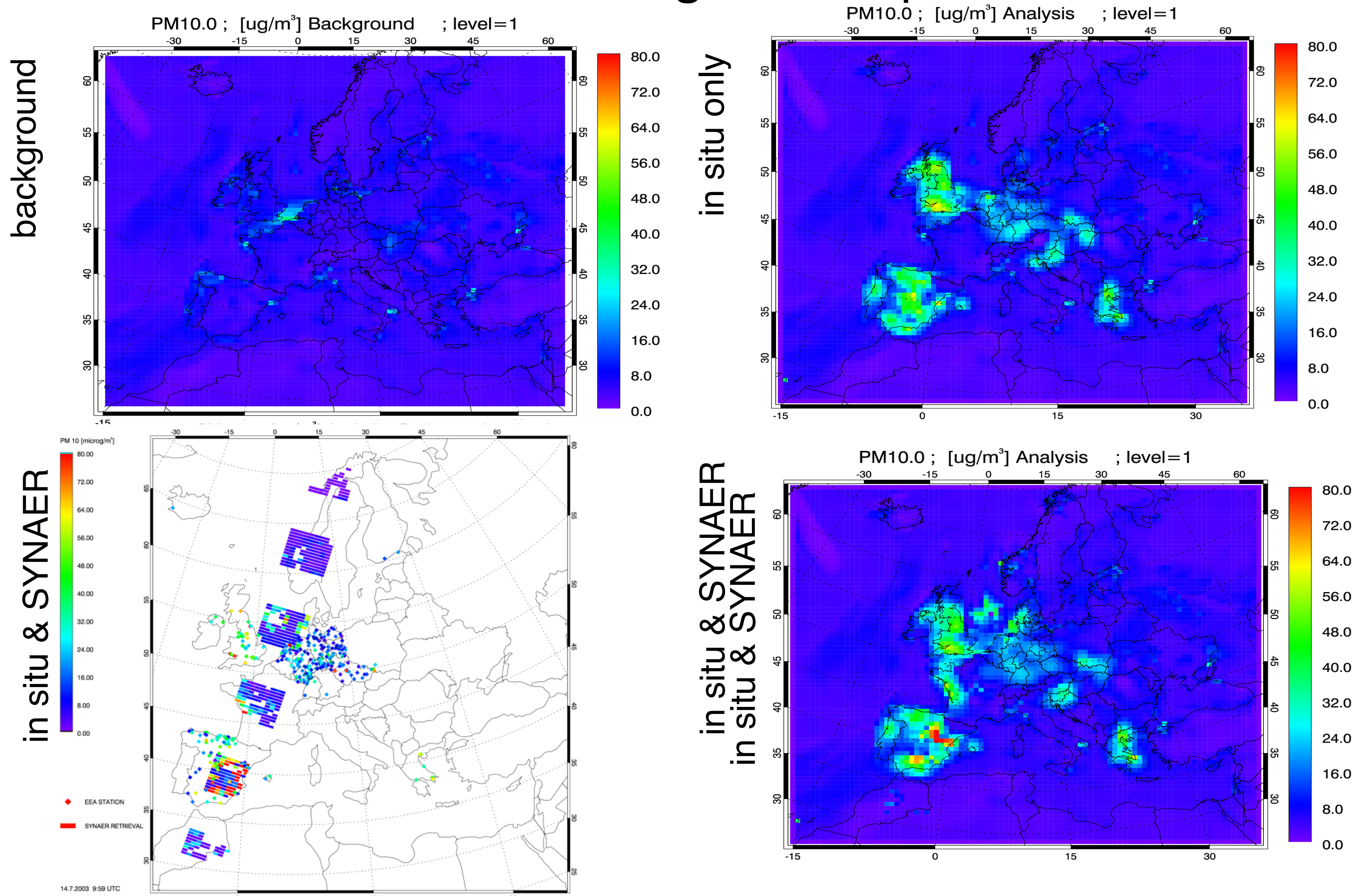
in situ & SYNAER



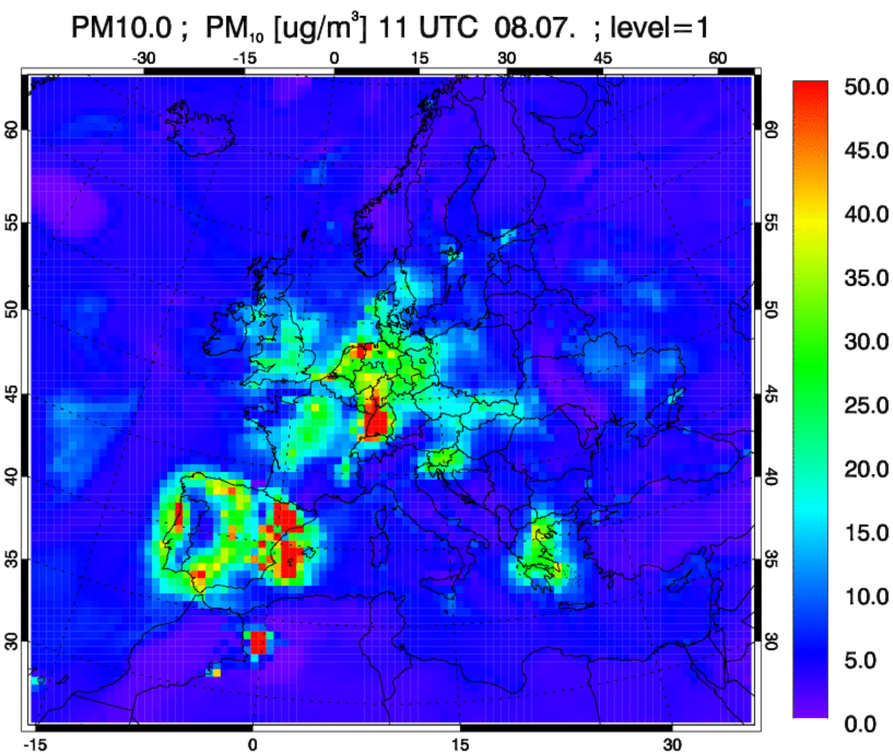
in situ & SYNAER



3Dvar aerosol assimilation (14.7.2003) biomass burning case in Spain

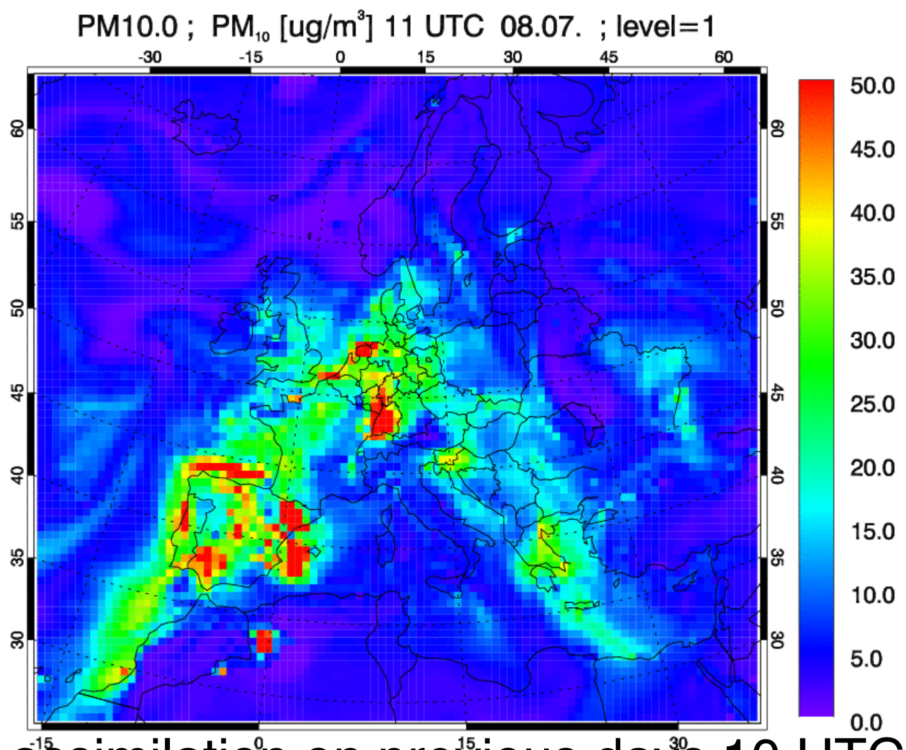


Do aerosol data assimilation effects accumulate? (14. July 2003)



No previous assimilation

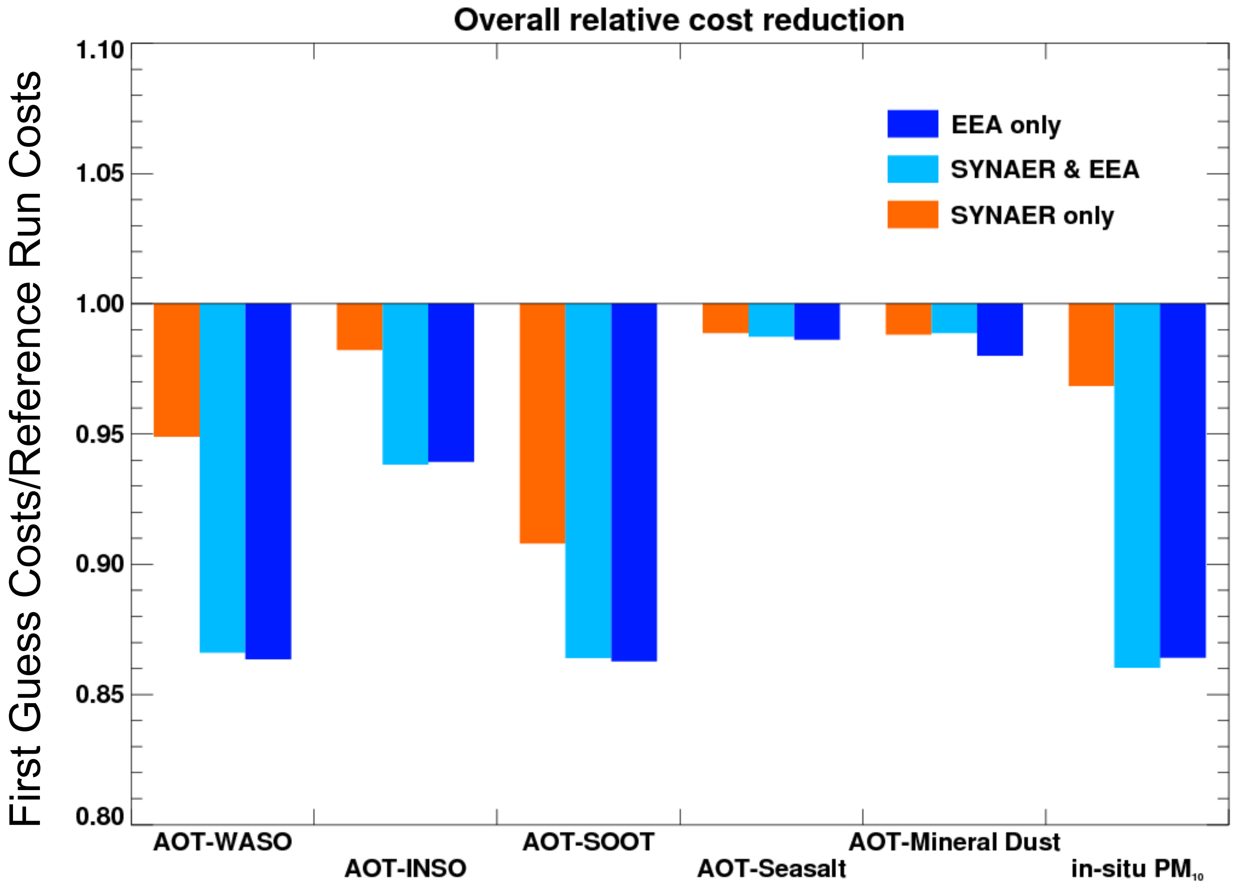
only 14. July 2003



assimilation on previous days 10 UTC

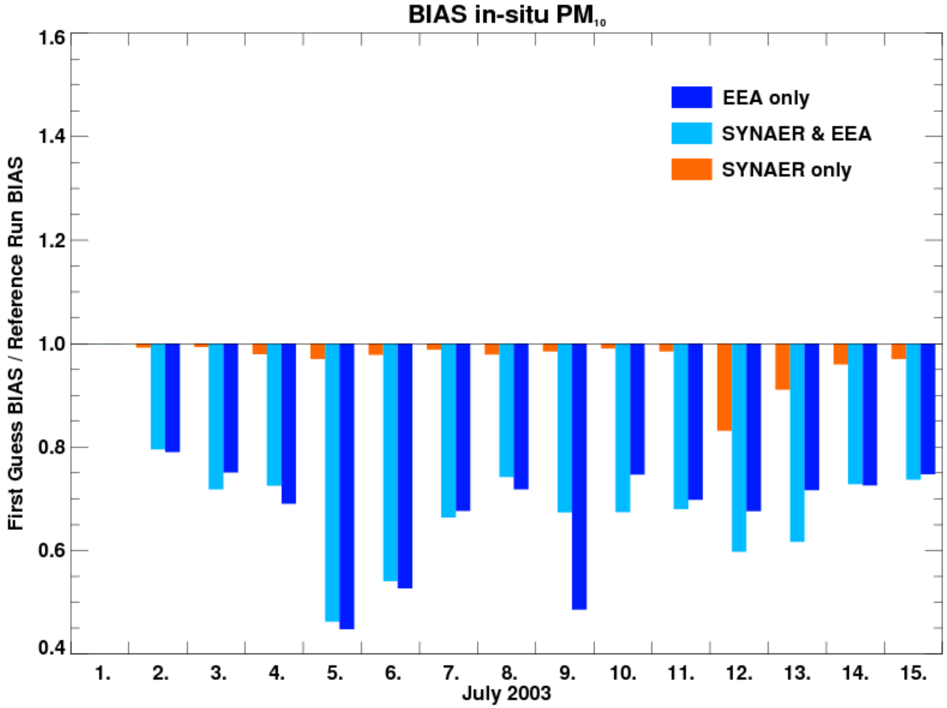
Accumulation of retrieval information over 14 days

Developement of forecast performance



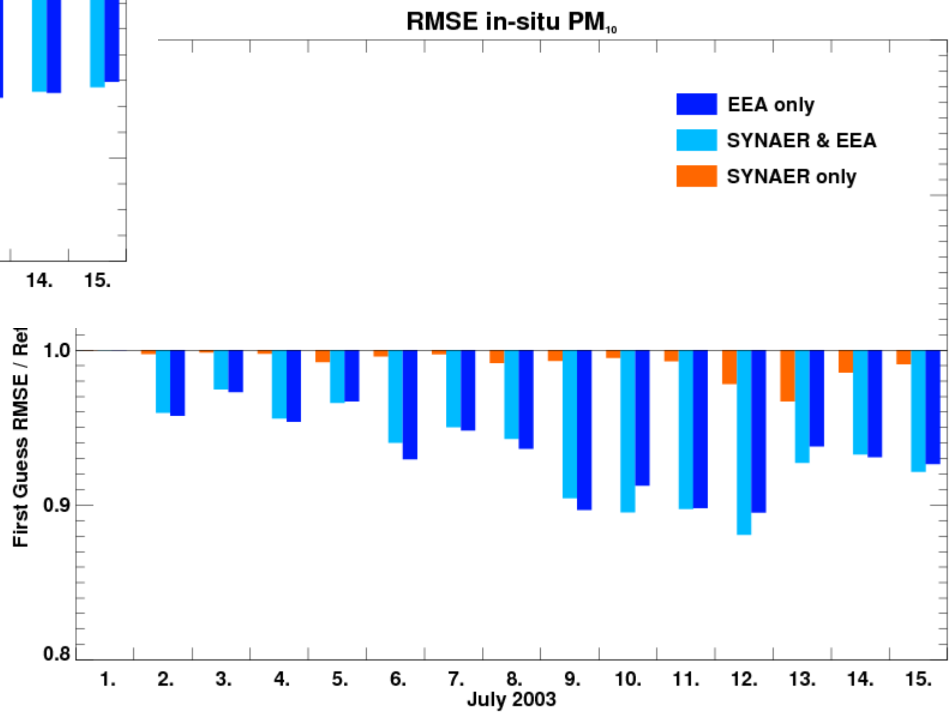
First Guess Costs / Reference Run Costs for measurements with a minimum relative change of 0.005.
SYNAER+EEA affects ~ 3% more observations than EEA

Developement of forecast performance



Overall BIAS reduction

SYNAER	0.96
SYN & EEA	0.67
EEA	0.67



Overall RMSE reduction

SYNAER	0.99
SYN & EEA	0.93
EEA	0.92

Case study II: ZEPTER-2 Campaign 2008

- Episode Oct 17 - Nov 8, 2008
- Assimilation acc. to flight schedule or 8 – 16/14/12 UTC
- Data assimilated
  - PND (Particle Number Densities) from Zepter2 – CPC
  - SYNAER-AOT
  - EEA in-situ PM_x
- Focus
  - Validate aerosol 4Dvar on nested grids
  - Assimilate PND as a new species

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

Model domains

EUR

- 107 x 97 x 23
- Resolution: 45 km
- Timestep: 600 s

CEN

- 70 x 76 x 23
- Resolution: 15 km
- Timestep: 240 s

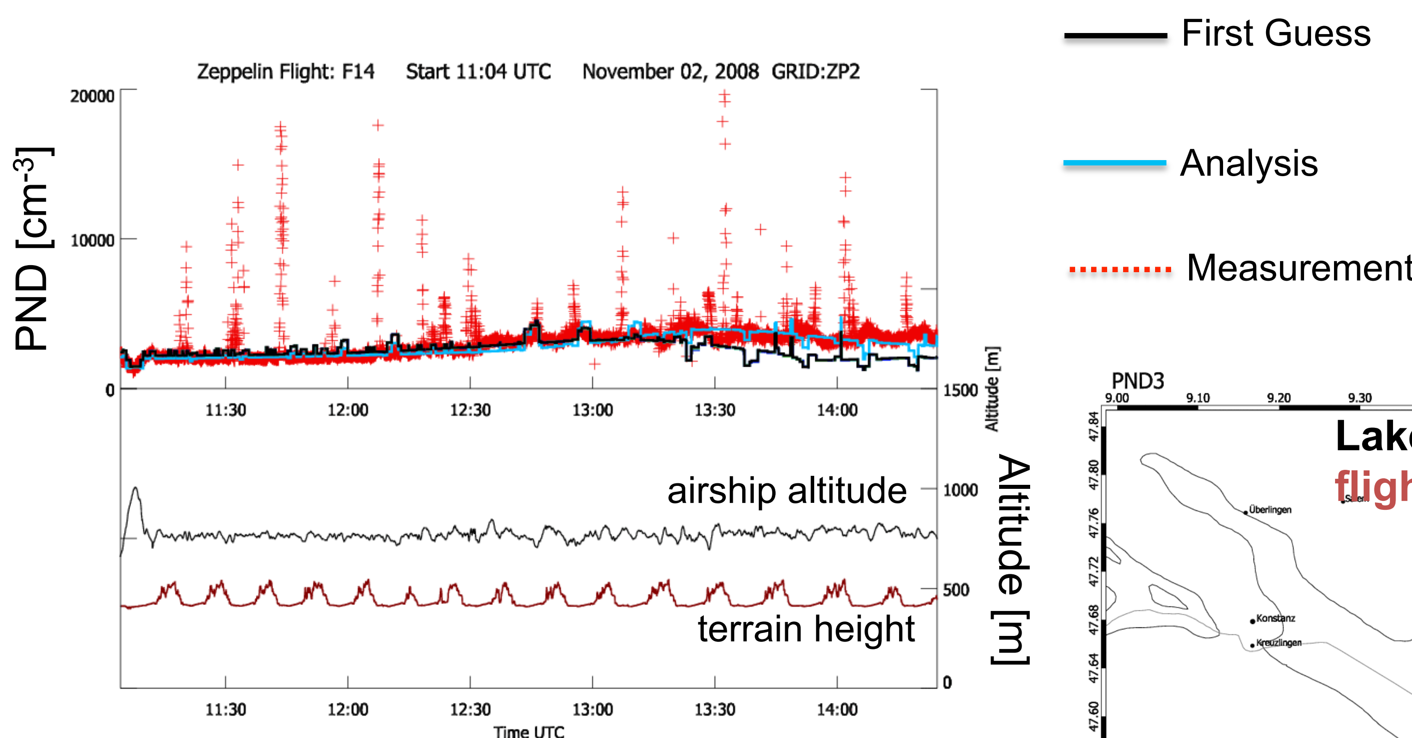
ZP2

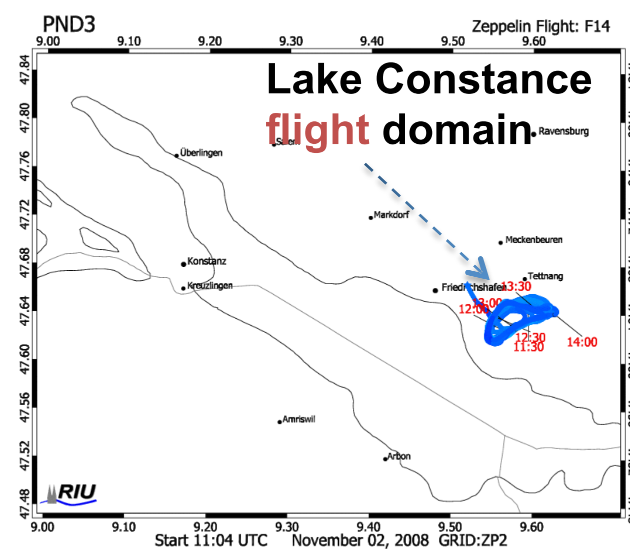
- 97 x 88 x 23
- Resolution: 5 km
- Timestep: 60 s

2. Analyses, Example (ii):

Zepter 2: 4D-var assimilation of particle number densities

Flight 14 assimilation of PND (0.005-3.0 µm) 02.11.2008 (11-15 UTC)



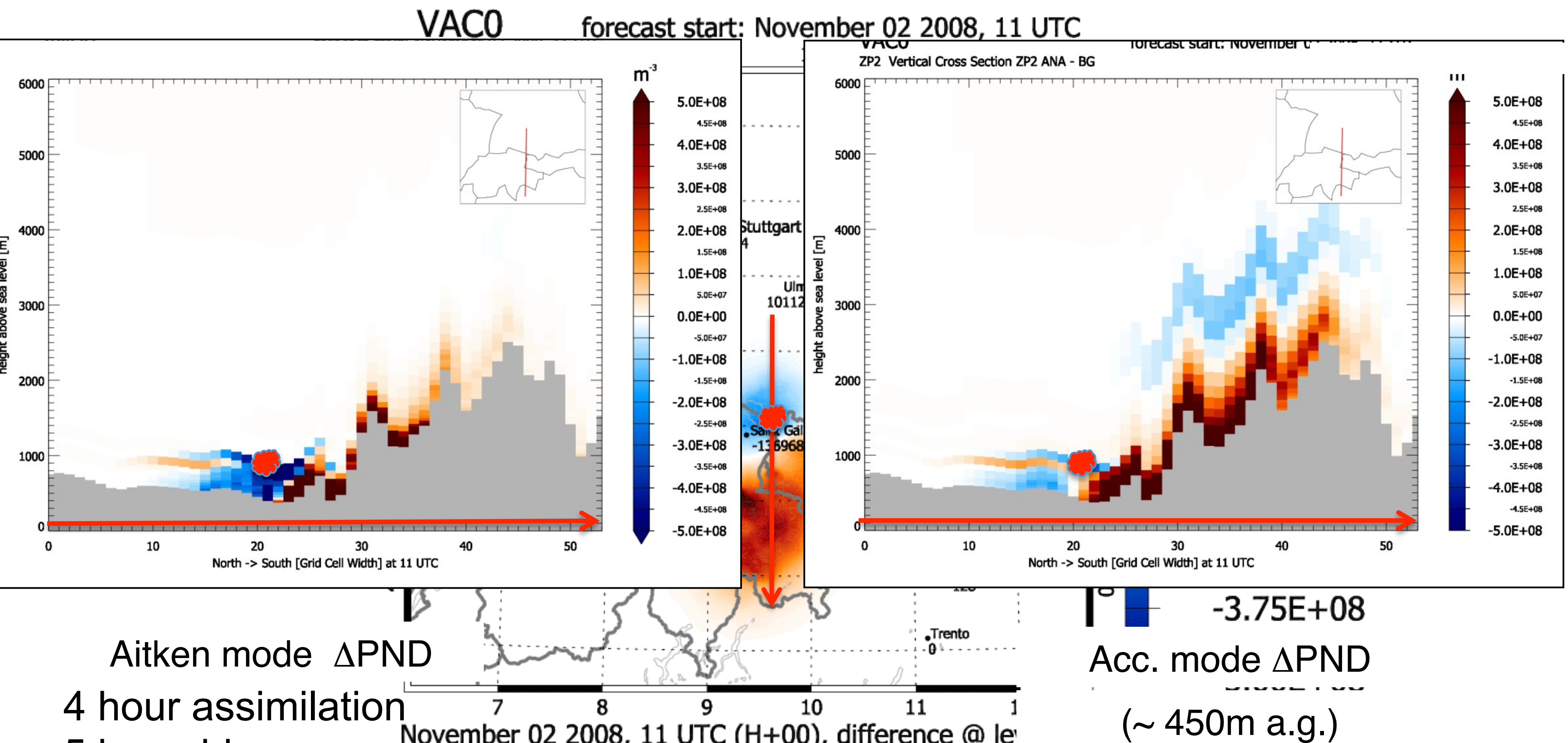


(from Lars Nieradzik, PhD thesis 2011)
water solubles include Friese and Ebel, 2010:

$$H^{+} - NH_4^{+} - Na^{+} - SO_4^{--} - NO_3^{-} - Cl^{-} - H_2O$$

2. Analyses, Example (ii cntd.):

Flight 14 assimilation of PND (0.005-3.0 µm), Nov. 2nd 2008



Aitken mode ΔPND

4 hour assimilation
5 km grid

Acc. mode ΔPND
(~ 450m a.g.)

from Lars Nieradzik, PhD thesis 2011

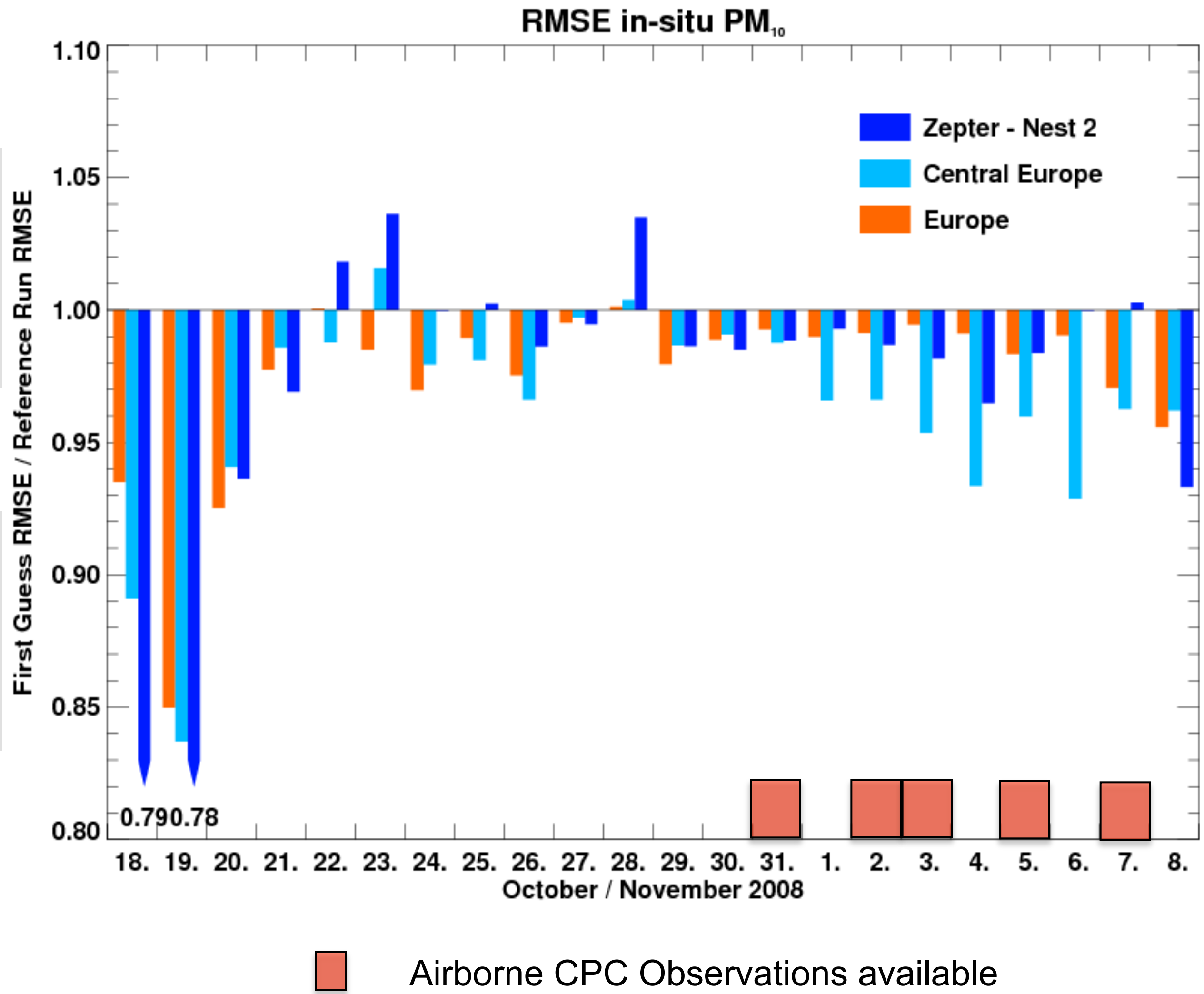
Development of forecast performance

Assimilation duration

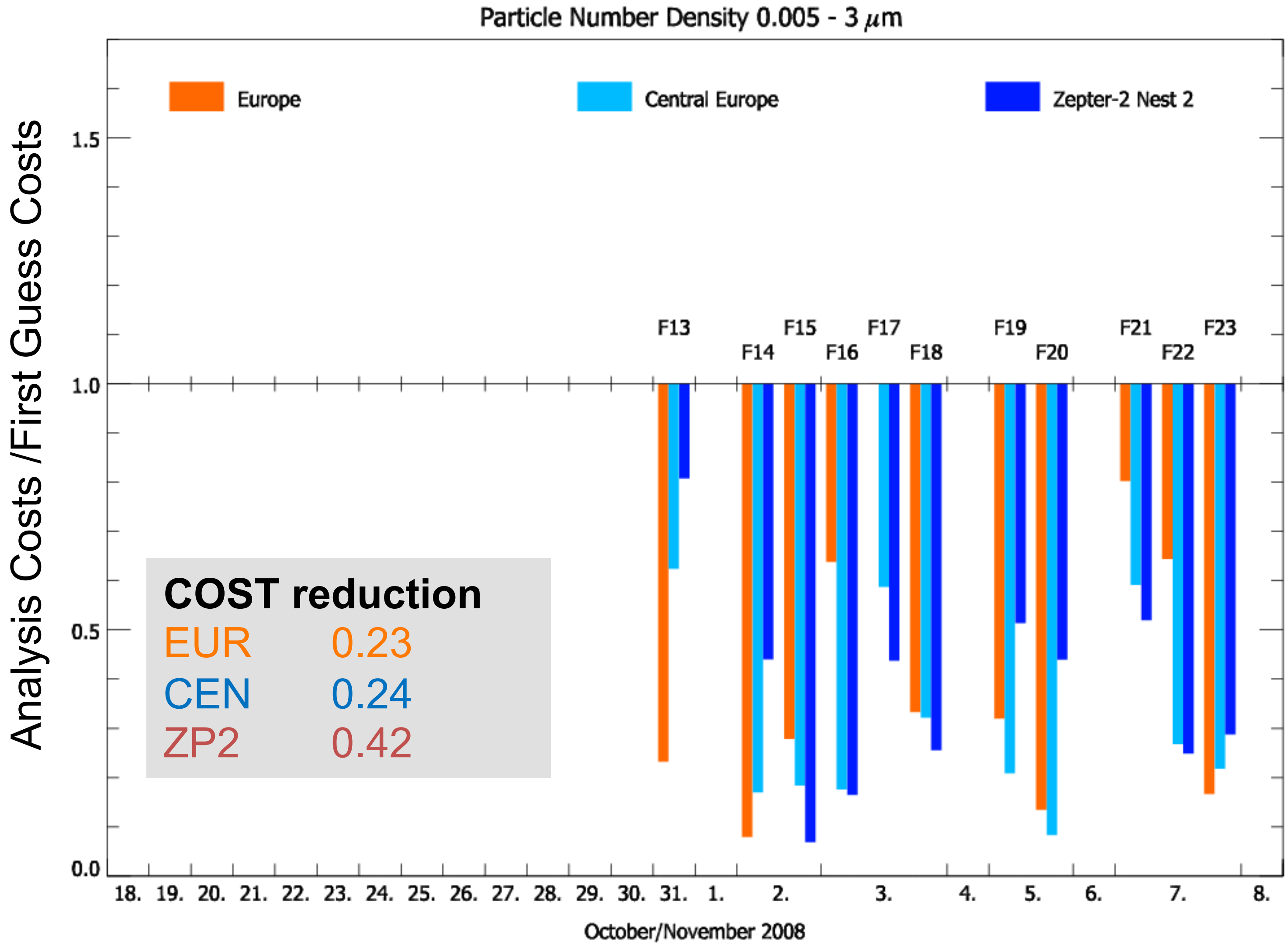
EUR 8 h
CEN 6 h
ZP2 4 h

RMSE reduction

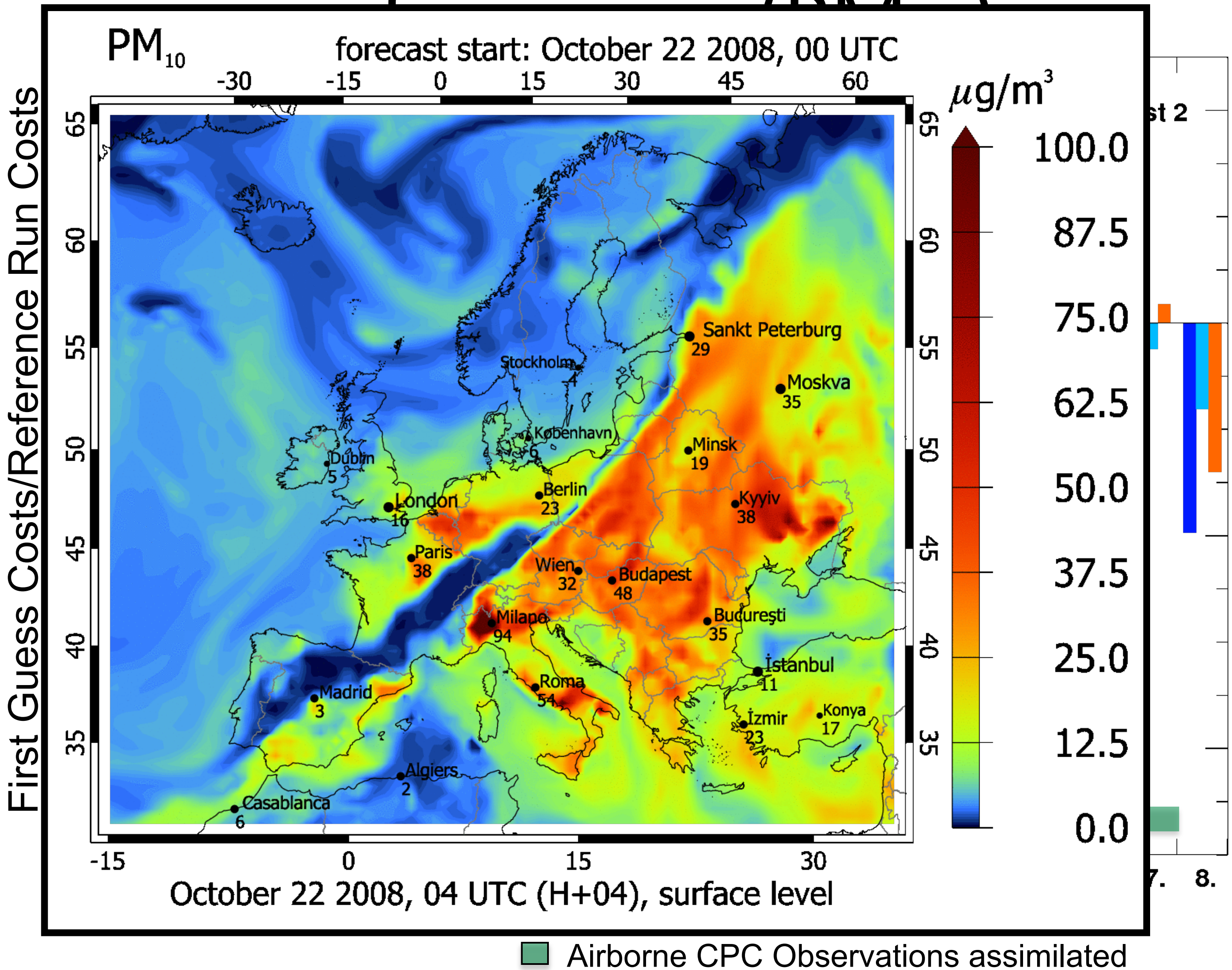
EUR 0.968
CEN 0.965
ZP2 0.971



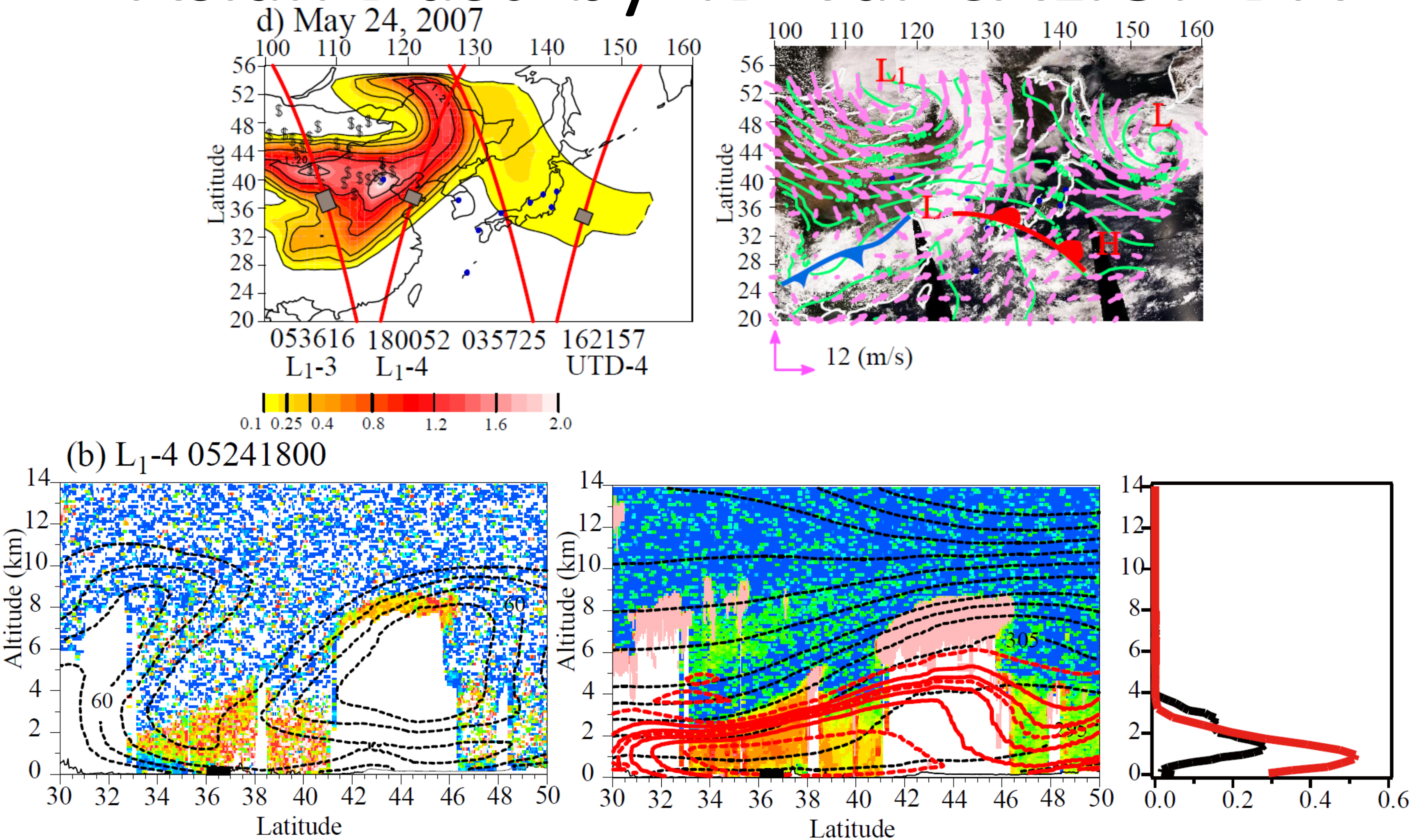
Assimilation statistics



Development of forecast

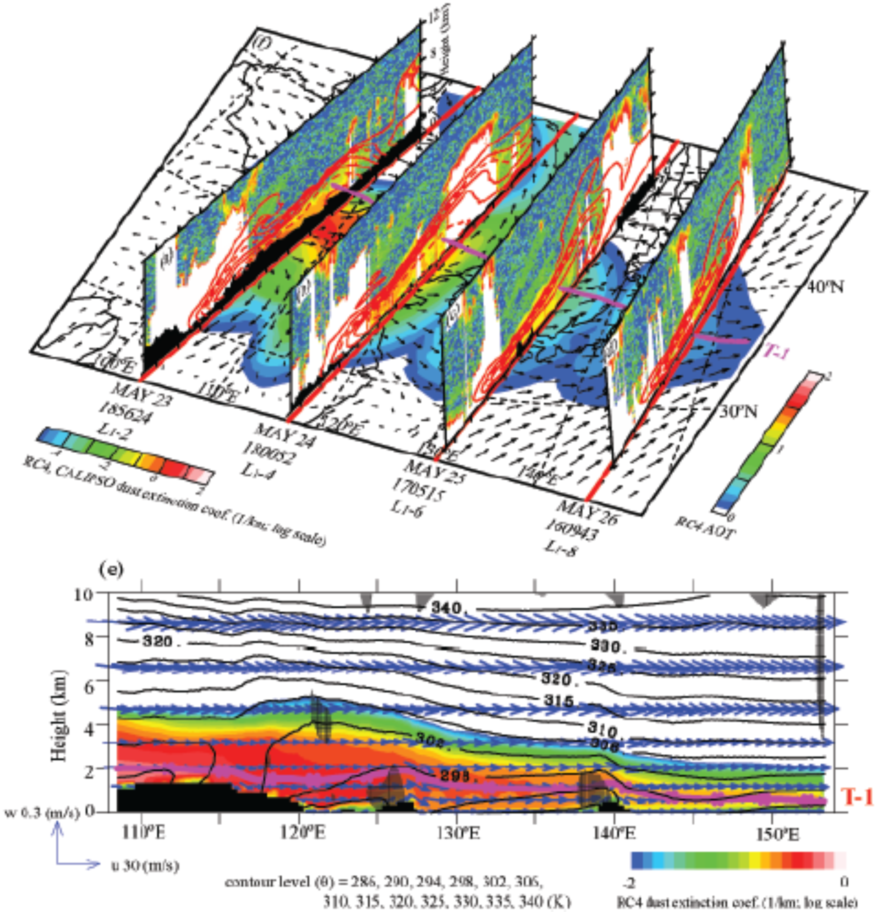


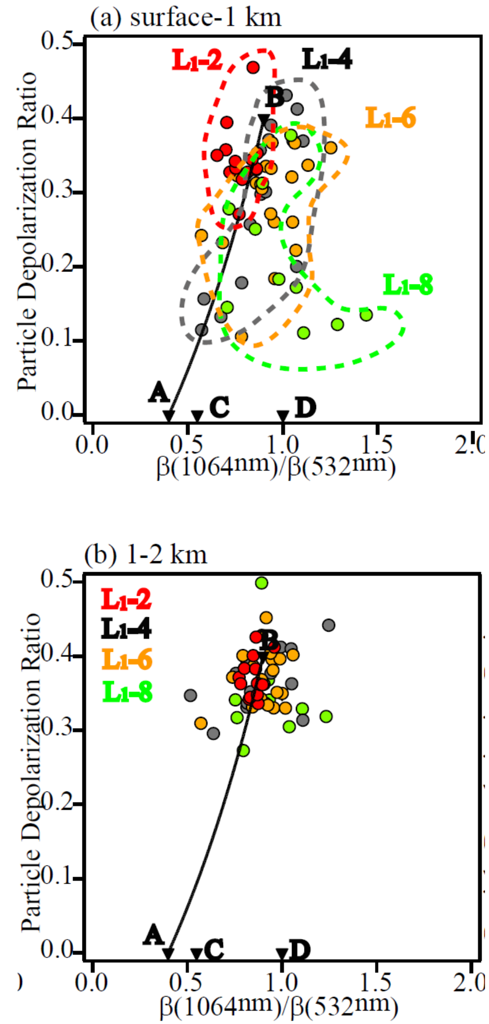
Asian Dust by 4D-var CALIOP DA



Y. Hara, K. Yumimoto, I. Uno, A. Shimizu, N. Sugimoto, Z. Liu, and D. M.Winker, 2009

Asian Dust by 4D-var CALIOP DA





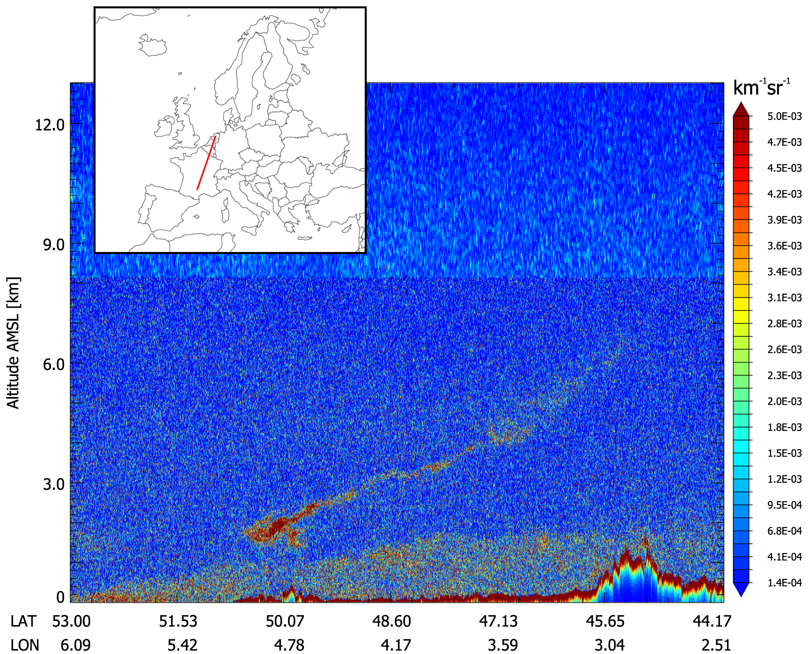
Y. Hara, K. Yumimoto, I. Uno, A. Shimizu, N. Sugimoto, Z. Liu, and D. M.Winker, 2009

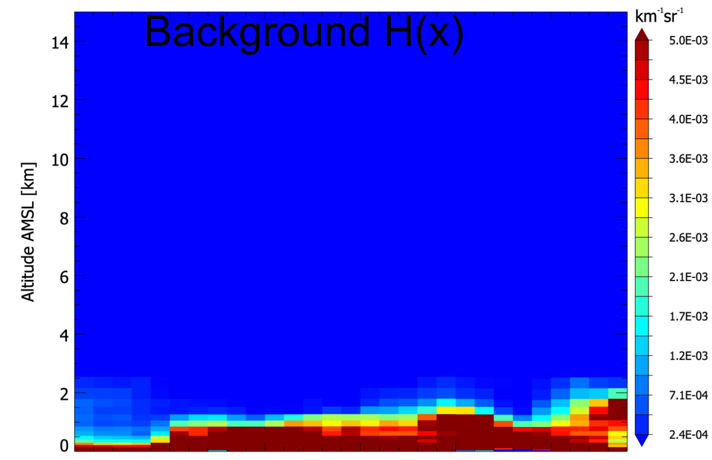
Example CALIOP
Variational volcanic ash data Assimilation Module
with selective background weakening for special events

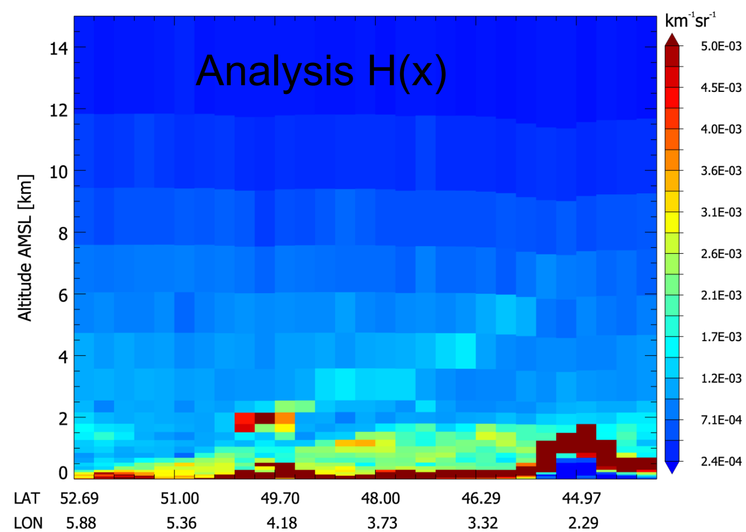
LiDAR 4D-var data assimilation for improved analysis of unexpected aerosol events
→ automated online adaptation of background error covariace matrix

CALIOP observation of the Eyjafjallajökull ash cloud

17 April 2010, 02:01:19 - 02:14:53 UTC (Winker et al., 2012)







A. Lange, master thesis

Summary

- chemical weather forecasts are a multiple scale problem
- chemical data assimilation rests on sparse and heterogeneous observations, with variable error characteristics (incl. error of representativity)
- **initial value optimisation is insufficient, as at least emissions are less known and more important**
- **the ability for inversion is therefore required to optimize emission rates**
- much is to be done for optimising multivariate covariance matrices