Emission rate and chemical state estimation by 4-dimensional variational inversion

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further contributions

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Structure of the presentation

1. Introduction: rationale of data assimilation for air quality inversion

2. Theory: critical implementation items

3. Results

4. Did we mature?

Motivations for tropospheric chemistry data assimilation efforts:

A means to fuse of heterogeneous data and information sources for

1. better predictability and sensitivity analyses
2. better estimation of principal parameters: e.g. emissions
3. better process simulations
4. better chemical state monitoring on regular grids; potentially earlier trend signal detection and attribution

Ad 2: Today's EPAs concerns
(Germany at least)

- sulphor, CO, ozone: considered under control

- PM10, PM 2.5: appropriate reduction measures are believed to be taken

- NO2: with new EC directive **no ideas as what strategy is promising**

Ad 4: Supernational initiatives

- **<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)**

Characteristics of tropospheric chemistry data assimilation (1) physical viewpoint

Main sources of uncertainty:

- direct parameters
  - Initial values, lateral boundary values
  - emission rates,
  - deposition and sedimentation velocities
  - reaction rates, J-values
- indirect parameters
  - boundary layer height
  - vertical exchange mechanisms: convection

Characteristics of tropospheric chemistry data assimilation (2), mathematical viewpoints

- highly underdetermined system - on 2 levels
  - variables/gridpoint: ~ 60 -200
  - satellite data: scalar column value → profile vector

- regionally/locally highly nonlinear chemical dynamics (photo chemistry)

- constraints by physical laws/models are insufficient, however central manifolds variable

- assimilation or inversion problem to be solved?

2. Theory: Critical implementation items

A) Inclusion of the emission inversion

B) Inhomogeneous Background error covariance formulation

C) Preconditioning

D) Implementation

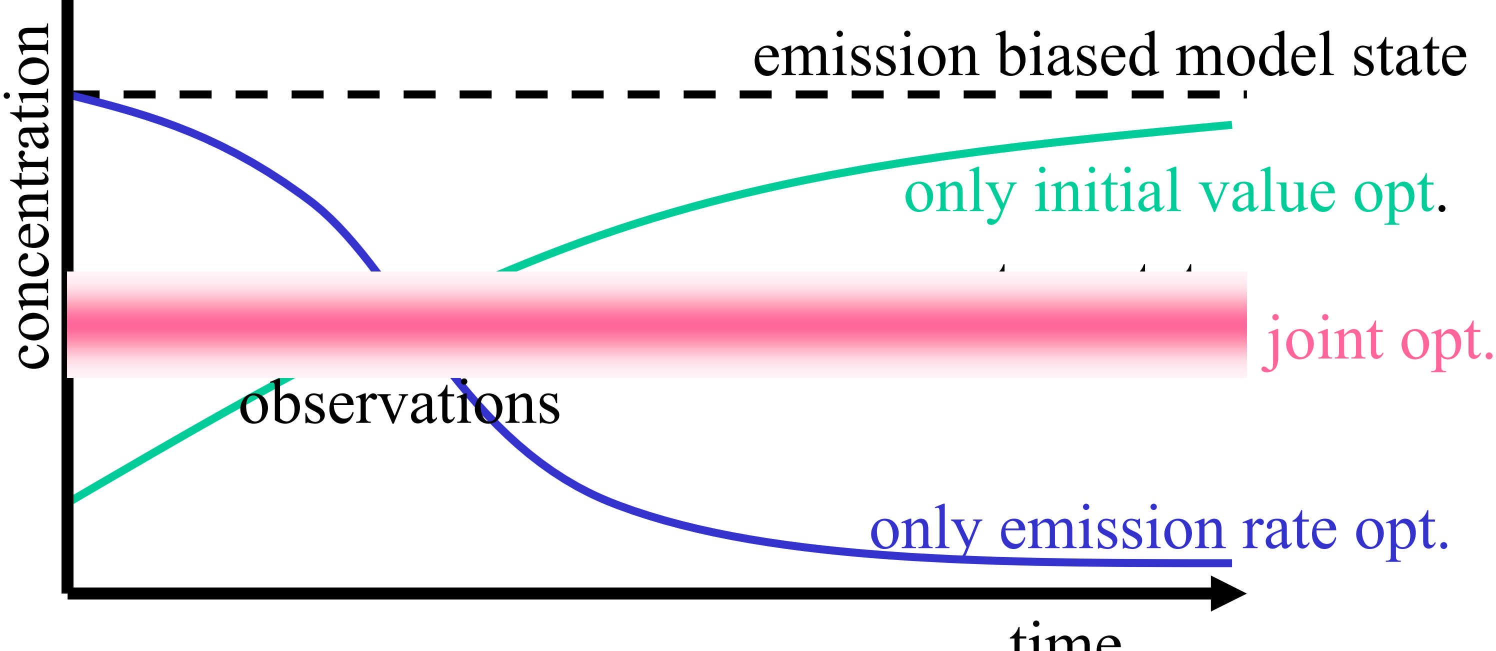
The most popular strategy: Linear estimation theory

- Provides for a
- Best Linear Unbiased Estimate (**BLUE**)

- However:
- assumes Gaussian error characteristics for positive semi-definite parameters by observations, forecasts, models

Question:
Which model parameter to be optimized by assimilation/inversion?

General Rule:
Parameter with maximal product
(impact on forecast skill) x (paucity of knowledge)
subject to some suitable norm x
Hypothesis:
initial state and emission rates are least known



The generalized cost function

$\mathcal{J}$ scalar functional on the time interval $t_0 \leq t \leq t_N$ dependent on $\delta\mathbf{x}(t)$

$$\boxed{\begin{aligned}&\mathcal{J}(\delta\mathbf{x}(t_0), \delta\mathbf{u}) = \\ &\frac{1}{2}(\delta\mathbf{x}(t_0))^T \mathbf{B}^{-1} \delta\mathbf{x}(t_0) + \boxed{\frac{1}{2}(\delta\mathbf{u})^T \mathbf{K}^{-1} \delta\mathbf{u}} + \\ &\frac{1}{2}\sum_{i=0}^{N} (\mathbf{d}(t_i) - \mathbf{H}(t_i)\delta\mathbf{x}(t_i))^T \mathbf{R}^{-1} (\mathbf{d}(t_i) - \mathbf{H}(t_i)\delta\mathbf{x}(t_i))\end{aligned}}$$

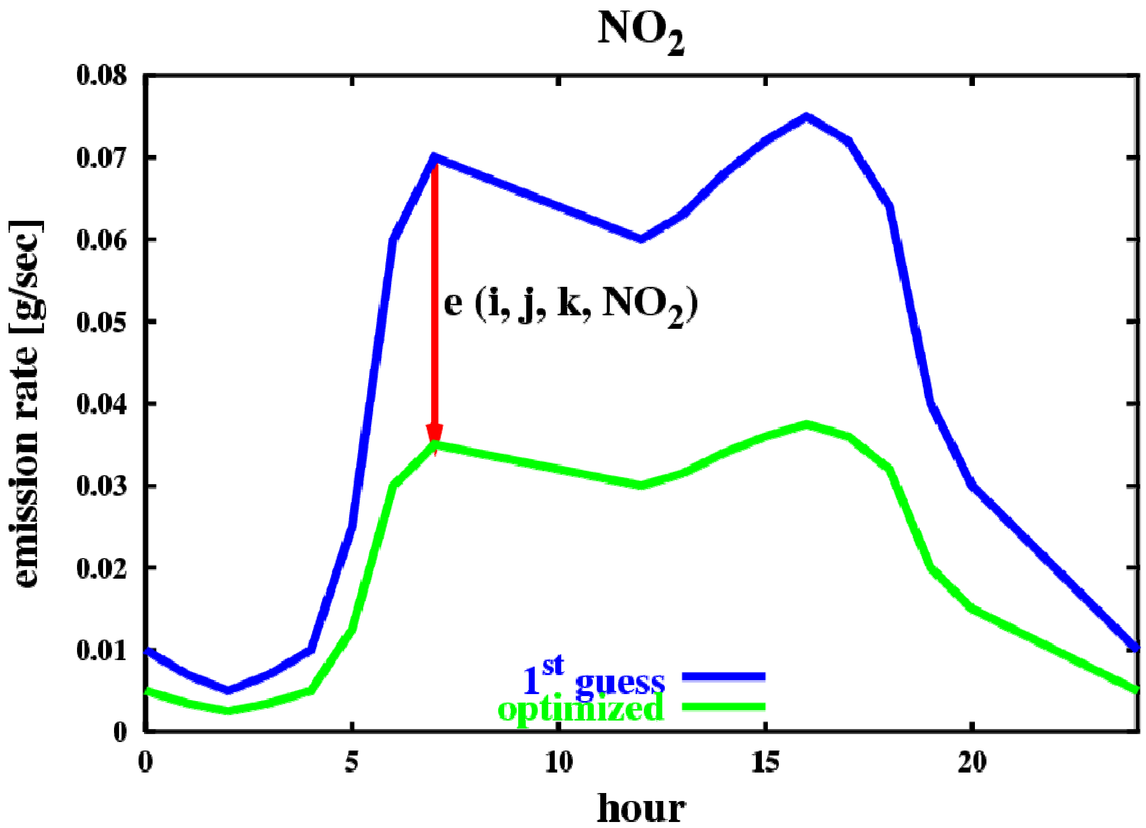
$\mathbf{H}(t) \in R^{M(t)\times N}$ is a linearised approximation of the forward observation operator $\mathcal{H}$.

The error covariance matrix of the background values $\mathbf{x}_b$ is $\mathbf{B}$
error covariance matrices of
emission perturbation $\delta\mathbf{u}$ is $\mathbf{K}$ and observation $\mathbf{d} = \mathbf{y} - \mathbf{x_b}$ is $\mathbf{R}$.

Optimisation of emission rates

diurnal emission profile as strong constraint

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 %

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 for preconditioning:

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

Computed by
diffusion approach, (see b
Cholesky factorisation

- Transformed cost Function:

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

- Transformed partial gradient:

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

Why special care for the background error covariance matrix **B**?

BECM can

- serve as hub for the distribution of ingested data
- balance weights of information from forecasts and information from observation/retrieval
- **distribute observation information spatially** and across variables
- serve as precondition information for minimisation

2 outstanding problems:

1. With linear estimation: How to treat the background error covariance matrix **B** (O(10^{14}))?
2. With variational methods: How can this be treated for preconditioning? (need $\mathbf{B}^{-1}$, $\mathbf{B}^{1/2}$, $\mathbf{B}^{-1/2}$)

Solution:

Diffusion Approach

Transformation of cost-function:

$$\boldsymbol{v} = \mathbf{B}^{-1/2}[\boldsymbol{x} - \boldsymbol{x}_b]$$

$$J(\boldsymbol{v}) = J_b + J_o = \frac{1}{2}\boldsymbol{v}^T\boldsymbol{v} + J_o$$

$$\nabla_{\boldsymbol{v}} J = \boldsymbol{v} + \mathbf{B}^{T/2}\nabla_{\boldsymbol{x}_0} J_o$$

$$\boldsymbol{x}_0 = \mathbf{B}^{1/2}\boldsymbol{v} + \boldsymbol{x}_b$$

minimisation procedure

=> Inverse of **B** and $\mathbf{B}^{-1/2}$ are not needed, if x_b= 1. guess.

Formulation of the background error covariance matrix: Diffusion paradigm

(Weaver and Courtier, 2001)

4D var needs the square root of the background error covariance matrix **B** ($\mathbf{O}$=10[12

Basic idea:

1. formulate covariances by Gaussians
2. approximate Gaussians by integration of the diffusion operator over time T
3. calculate $B^{1/2}$ by integration over time T/2 (comp. cheap), and
4. intermittent normalisation (comp. more challenging)

$$\mathcal{C} : \eta(z,0) \rightarrow (4\pi\kappa T)^{1/2}\eta(z,T)$$

with

$$\eta(z,T) = (4\pi\kappa T)^{-1/2} \int_{z'} \exp\left(-\frac{(z-z')^2}{4\kappa T}\right) \eta(z',0)dz'$$

and radius of influence

$$L^2 = 4\kappa T$$

$\mathbf{B}^{1/2}$ and $\mathbf{B}^{T/2}$ describing a quasi Gaussian correlation can be modelled using a diffusion operator:

$$\mathbf{B} = \mathbf{B}^{1/2}\mathbf{B}^{T/2} = \left(\boldsymbol{\Sigma\Lambda}\mathbf{L}^{1/2}\mathbf{W}^{-1/2}\right)\ \left(\mathbf{W}^{-1/2}\mathbf{L}^{1/2}\boldsymbol{\Lambda\Sigma}\right)$$

$\boldsymbol{\Sigma}$: Matrix of background error variances (diagonal)

$\boldsymbol{\Lambda}$: Matrix of normalisation factors (diagonal)

$\mathbf{L}$: Diffusion Operator

$\mathbf{W}$: Matrix of grid cell area elements (diagonal)

Background Error Covariance Matrix B

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

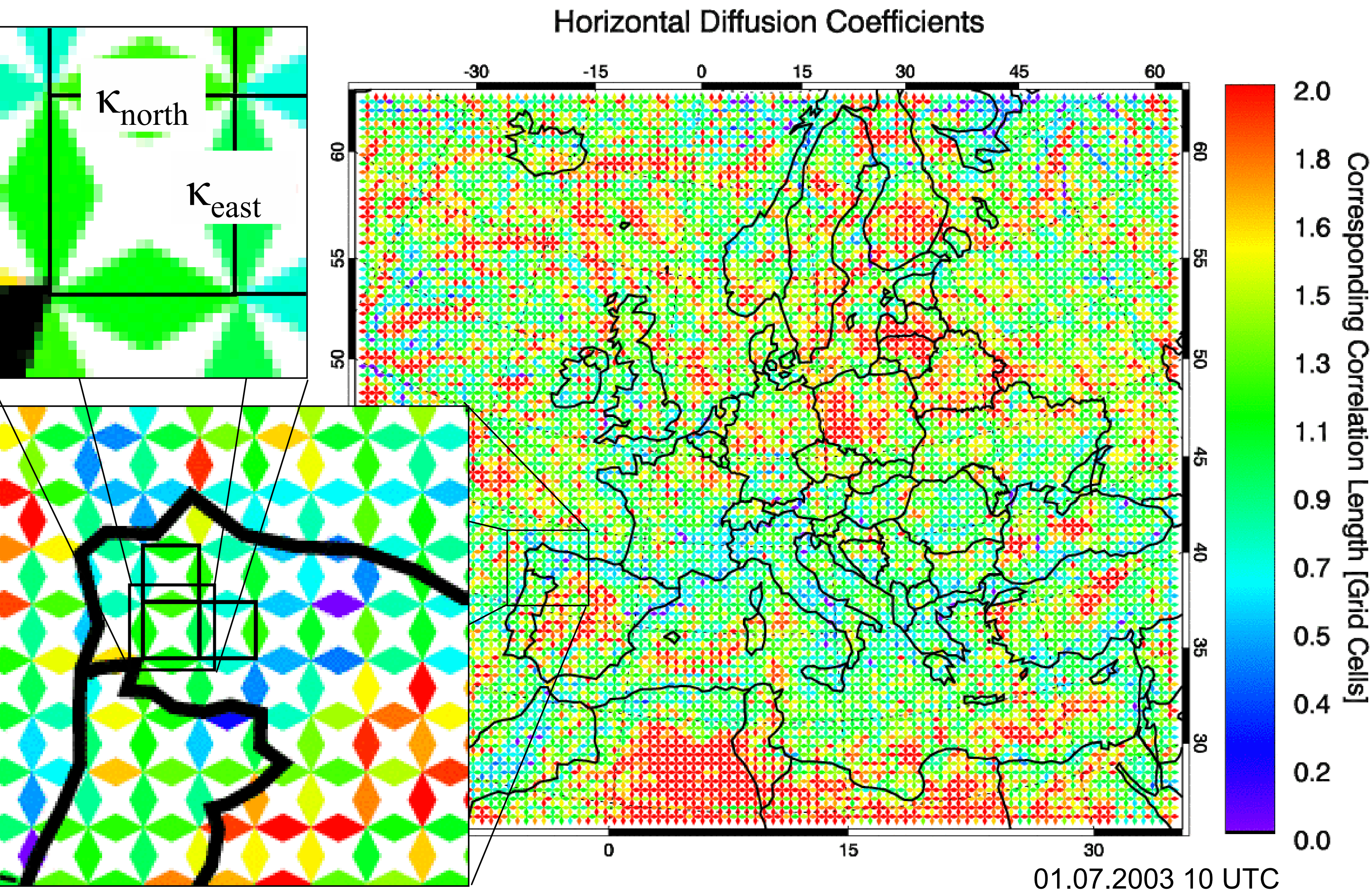
$$\mathbf{B} = \Sigma\,\mathbf{C}\,\Sigma$$

$$\mathbf{C} = \mathbf{C}^{1/2}\mathbf{C}^{\mathrm{T}/2}$$

$$\mathbf{C}^{1/2} = \mathbf{\Lambda}\Pi_N(\mathbf{L}_v^{1/2N}\mathbf{L}_h^{1/2N})$$

$$\mathbf{C}^{\mathrm{T}/2} = \Pi_N(\mathbf{L}_h^{\mathrm{T}/2N}\mathbf{L}_v^{\mathrm{T}/2N})\mathbf{\Lambda}$$

$\Pi(L_v L_h)$ approximates commutativity vertical and horizontal diff. op.


Horizontal Diffusion Coefficients
κ_{north}
κ_{east}
-30
-15
0
15
30
45
60
0
15
30
60
55
50
45
40
35
30
2.0
1.8
1.6
1.5
1.3
1.1
0.9
0.7
0.5
0.4
0.2
0.0
Corresponding Correlation Length [Grid Cells]
01.07.2003 10 UTC

EURAD-IM
4D-var system (1)

EURAD-IM adjoints

- RACM chemistry mechanism
- implicit vertical diffusion
- explicit horizontal diffusion
- Bott 4th order advection
- emissions: EMEP, TNO

MADE, SORGAM adjoint version under way

EURAD-IM
4D-var system (2)

- horizontal and vertical covariances: full anisotropy and inhomogeneity available by diffusion approach (Weaver and Courtier, 2001)
- preconditioning: options logarithmic, square root diffusion operator
- minimisation quasi-Newton by L-BFGS

Distribute observation information spatially

- **B** formally is of order O(**B**) ~ $(10^7)^{2:}$
- not tractable in practice, by volume and by information needs
- seek for a low dimensional control parameterization
- exploit external information (e.g. meteorological data, surface information (GIS))

Background Error Covariance Matrix B
(short design outline)

1. How to obtain the covariances?

K=# Ensembles; i,j neighboring cells

Ensemble/NMC approach:

$$\boxed{B_{ij} = \frac{1}{K}\sum_{n=1}^{K}\left(x_i^n - \overline{x}_i\right)\left(x_j^n - \overline{x}_j\right)}$$

2. How to process this information?

Translate into Diffusion coefficients→difusion paradigma

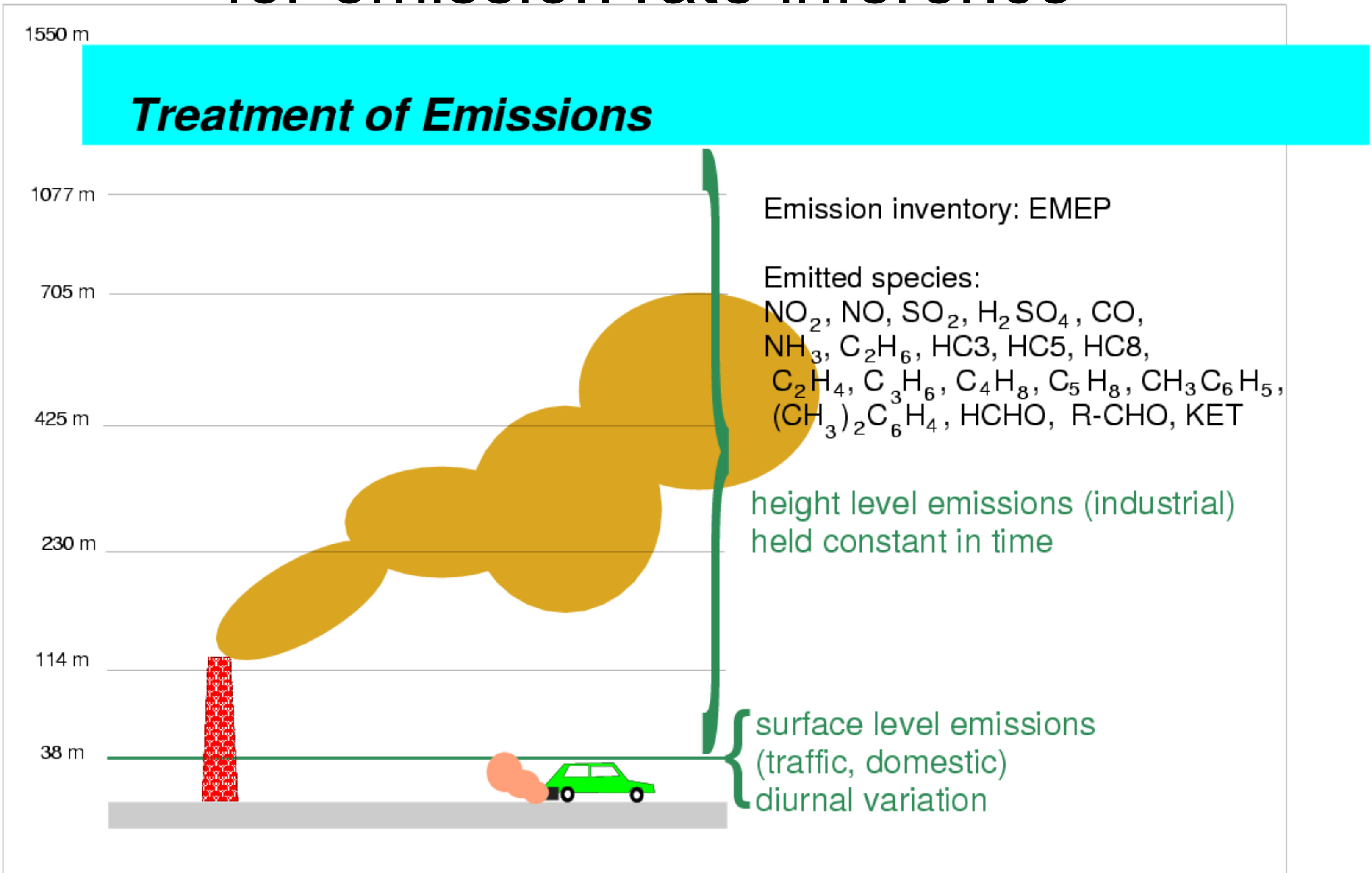
Correlation length L to neighboring gridcell:

$$B(r) = B(0)\exp\left(-\frac{r^2}{2L^2}\right), r = 1, B(1) = B_{ij}, B(0) = 1/2(B_{ii} + B_{jj})$$

$$\Rightarrow \boxed{L = \left(2\ln\left(\frac{B(0)}{B(1)}\right)\right)^{-1/2}}$$

diffusion coefficients κ: $L = \sqrt{2\kappa T}$

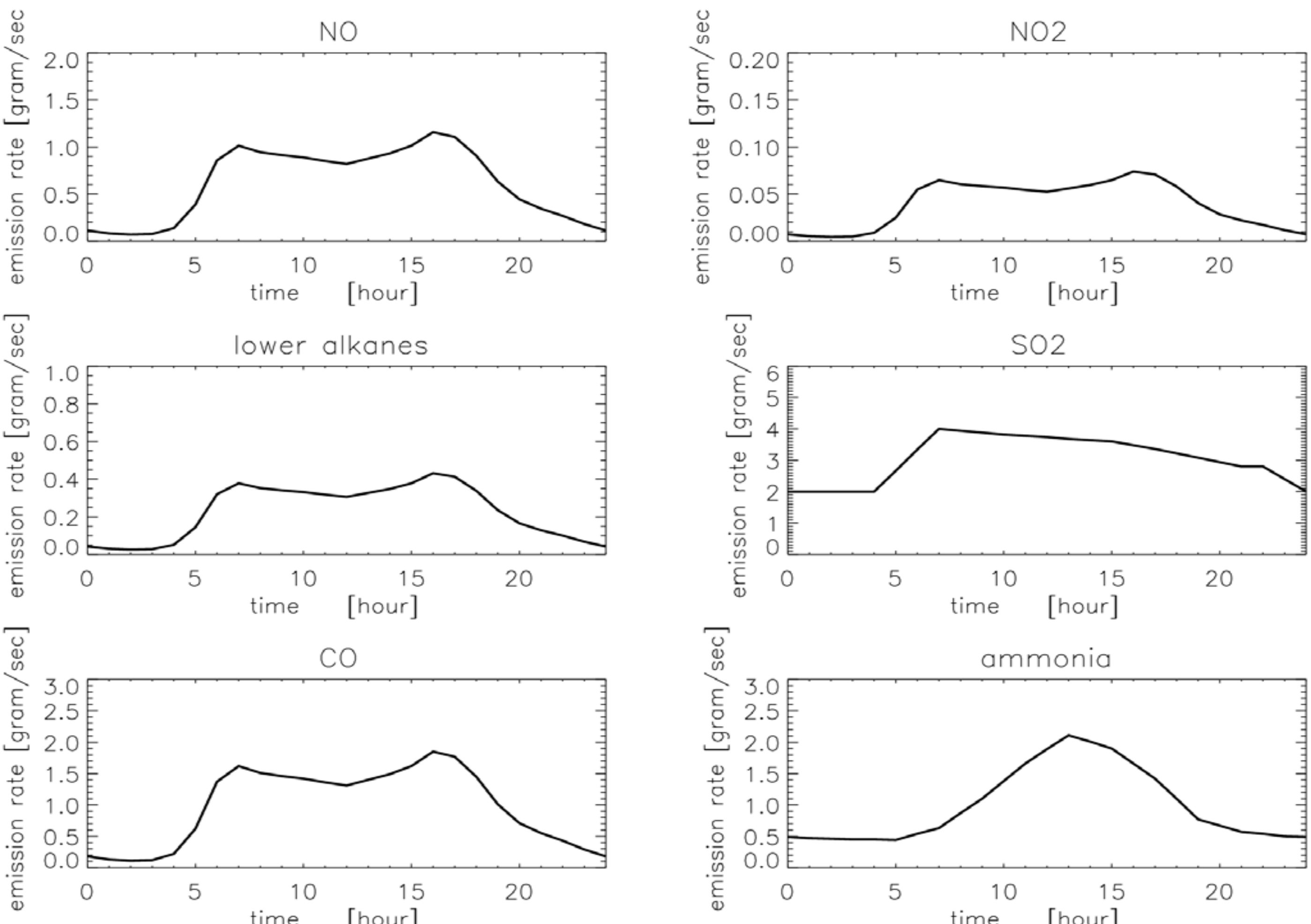
Treatment of the inverse problem for emission rate inference



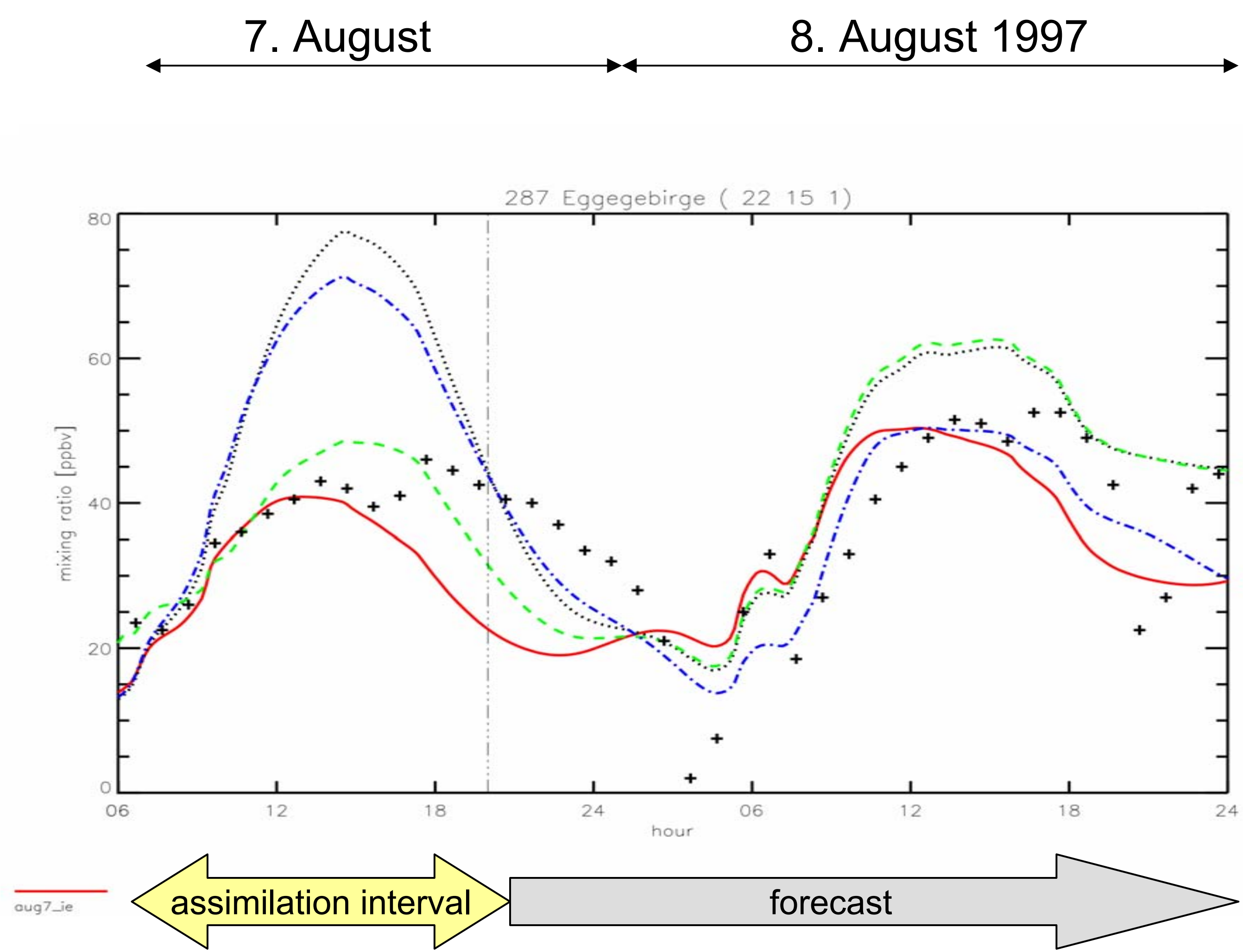
Normalised diurnal cycle of anthropogenic surface emissions *f(t)*

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

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



Semi-rural measurement site **Eggegebirge**



+ observations
no optimisation

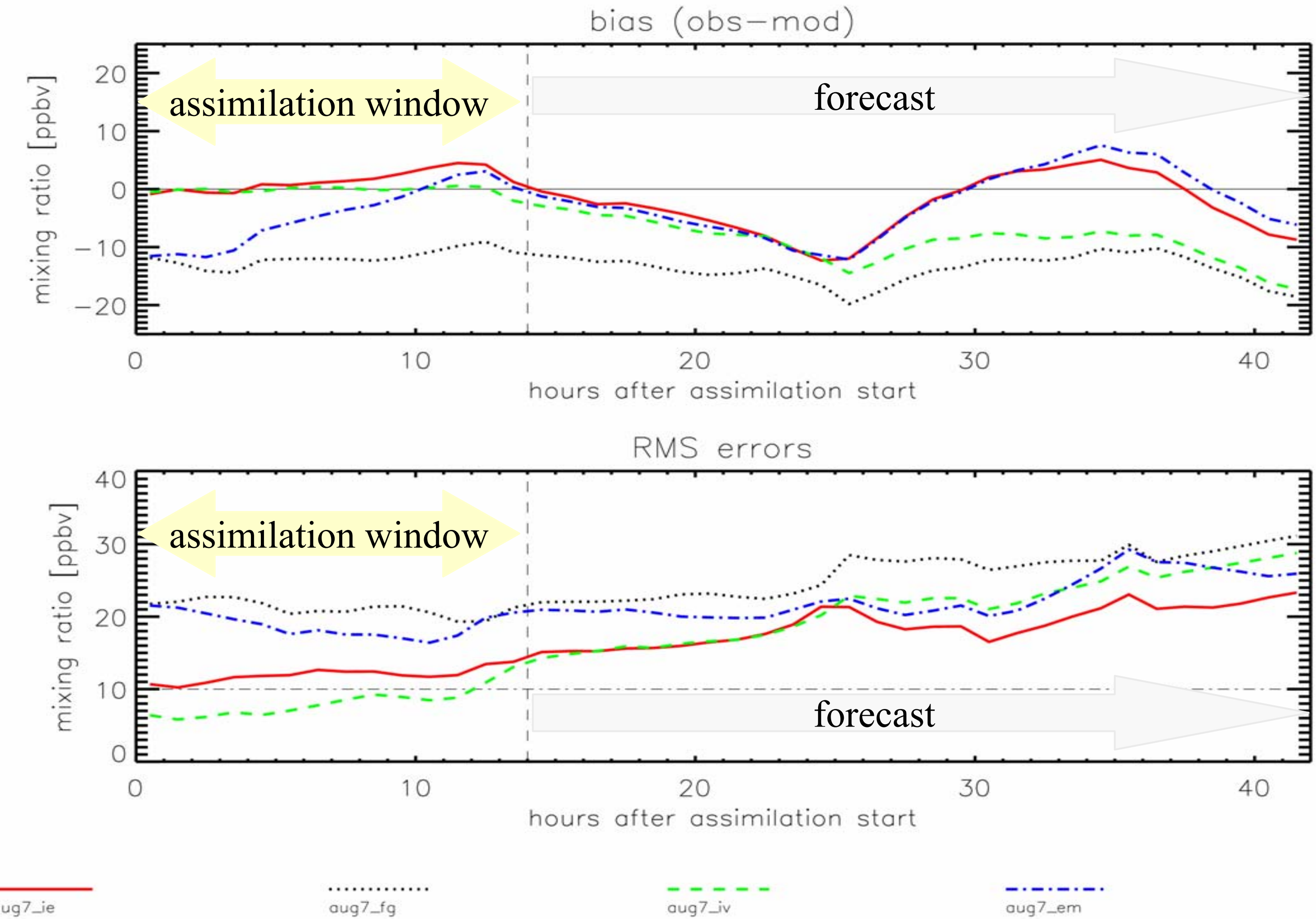
initial value op

emis. rate opt.

joint emis +
ini val opt.

(Elbern et al. 200

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



+ observations no optimisation

initial value op

emis. rate opt.

joint emis + ini val opt.

(Elbern et al. 200

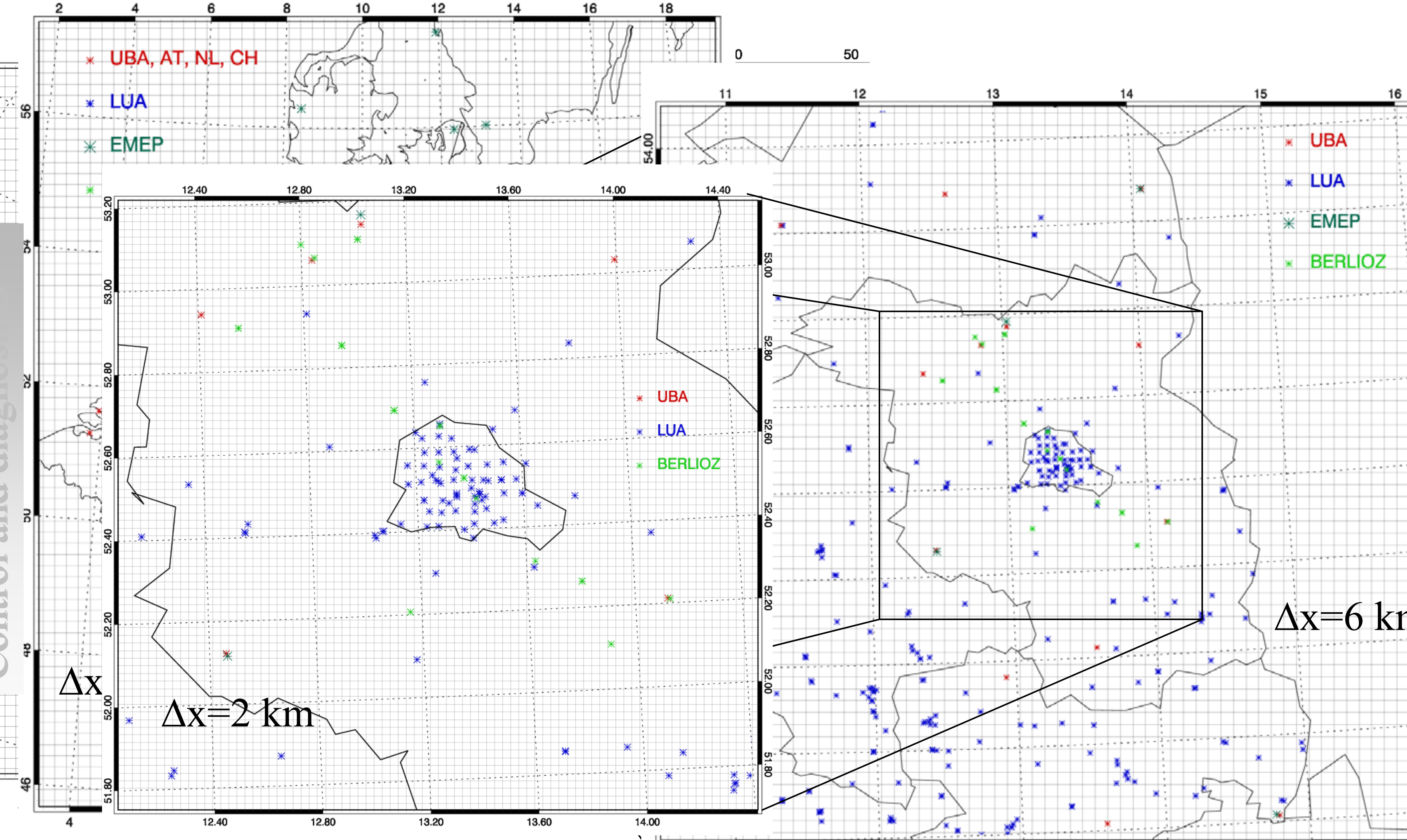
NO_x (=NO, NO_2) assimilation problems:

- NOx highly reactive (photochemistry
- observation site network not representative in most cases: bias toward urbanized areas
- routinely operated molybdenum converters notoriously sample much of NO_y (HNO3, PAN,…)
- satellite data as tropospheric columns with sensitivities unfavourable for the surface

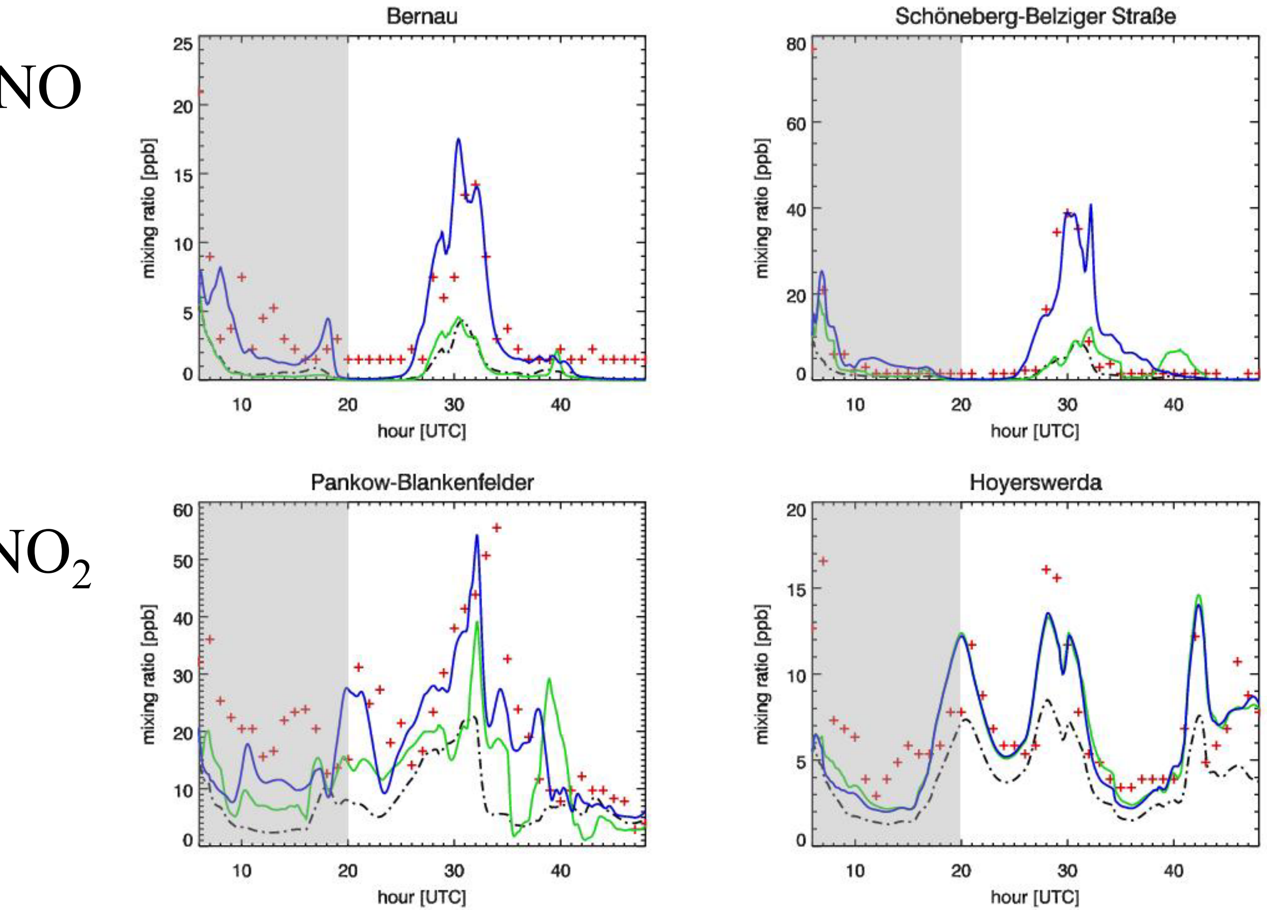
Which is the requested resolution?

BERLIOZ grid designs and observational sites

(20.→21. 07.1998)

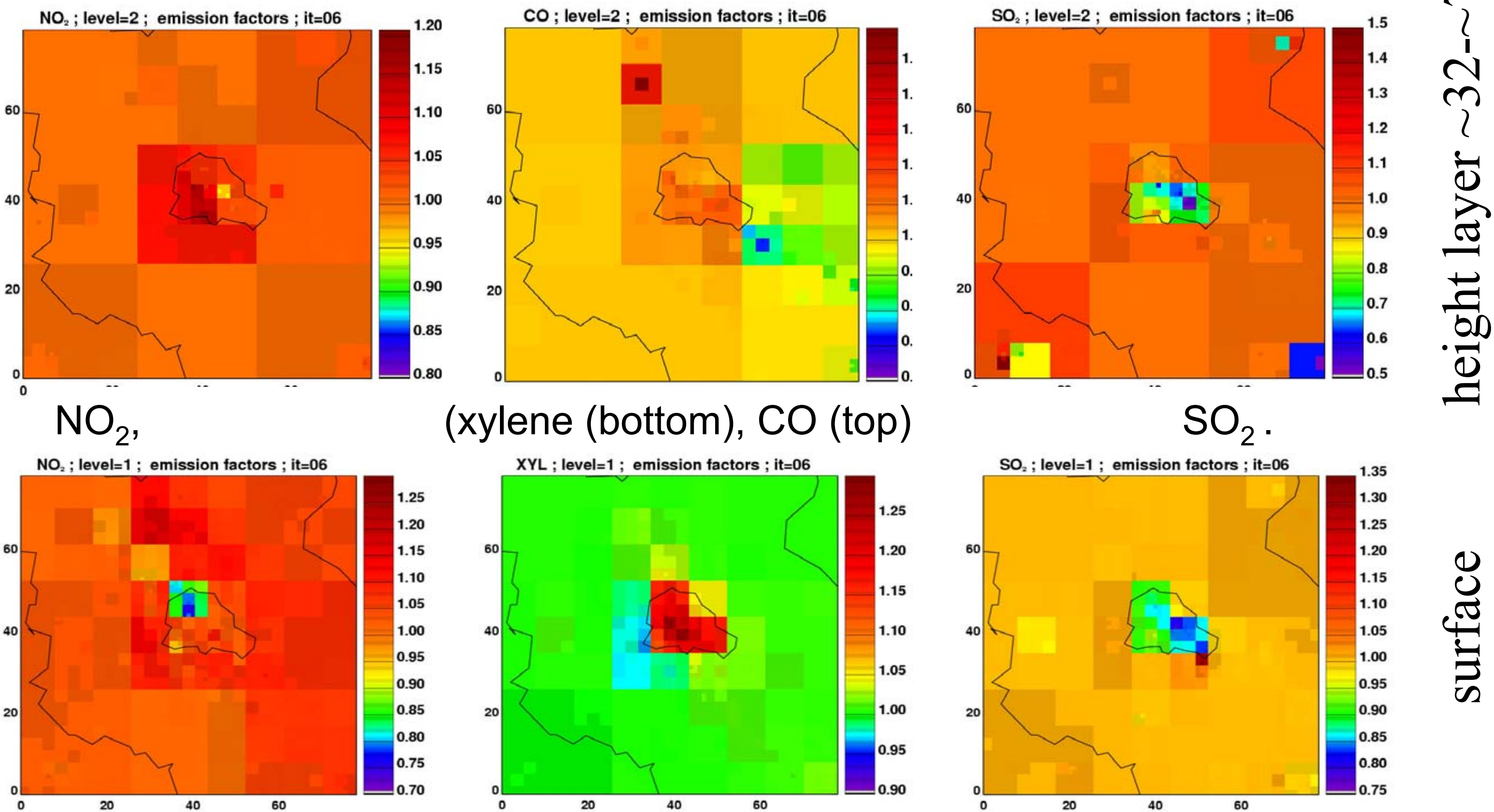


Some BERLIOZ examples of NOx assimilation (20.→21. 07.1998)



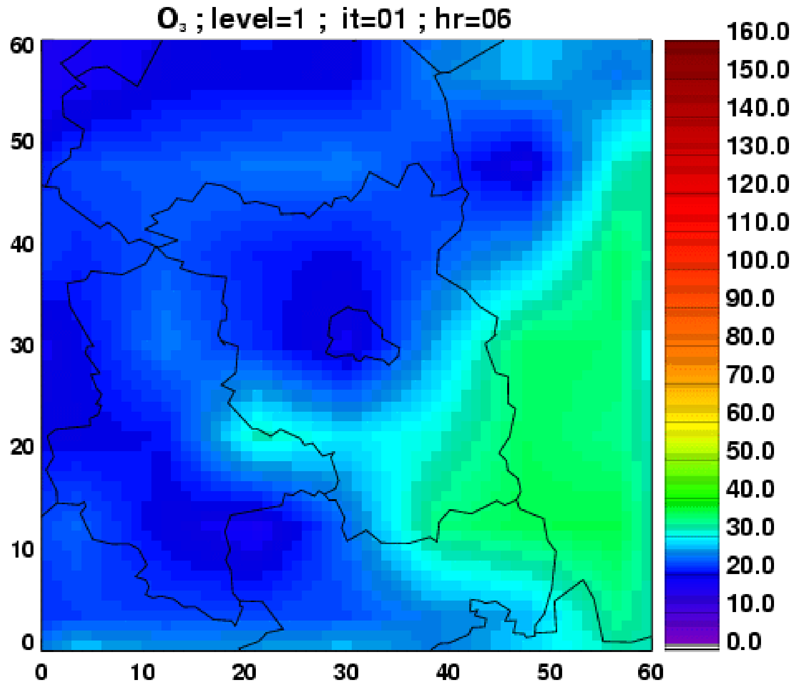
Time series for selected NO stations on nest 2.
+ observations,
-- - no assimilation,
-_____ N1 assimilation (**18 km**)
-____N2 assimilation.(**6 km**)
-grey shading: assimilated observations, others forecasted.

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

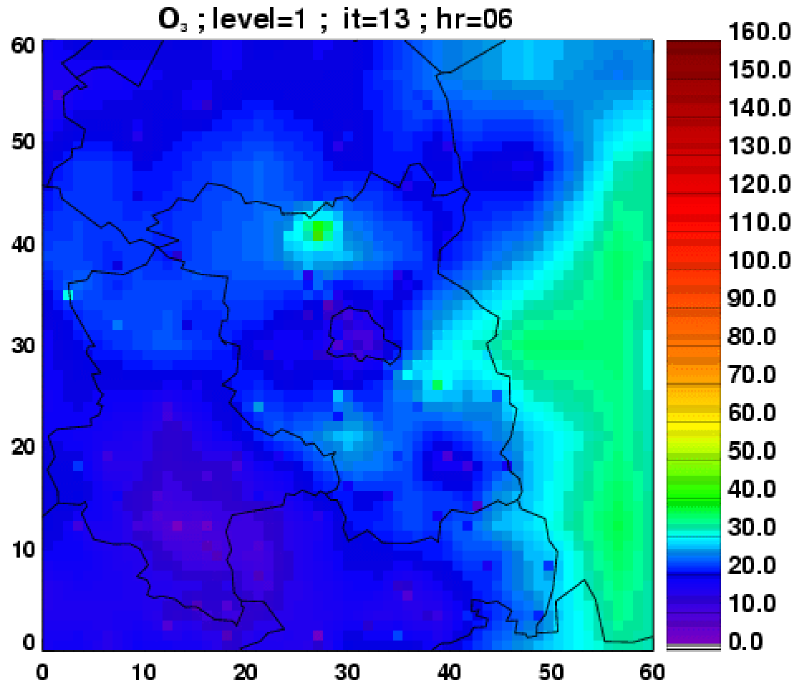


Nest 2: (surface ozone)
(20.→21. 07.1998)

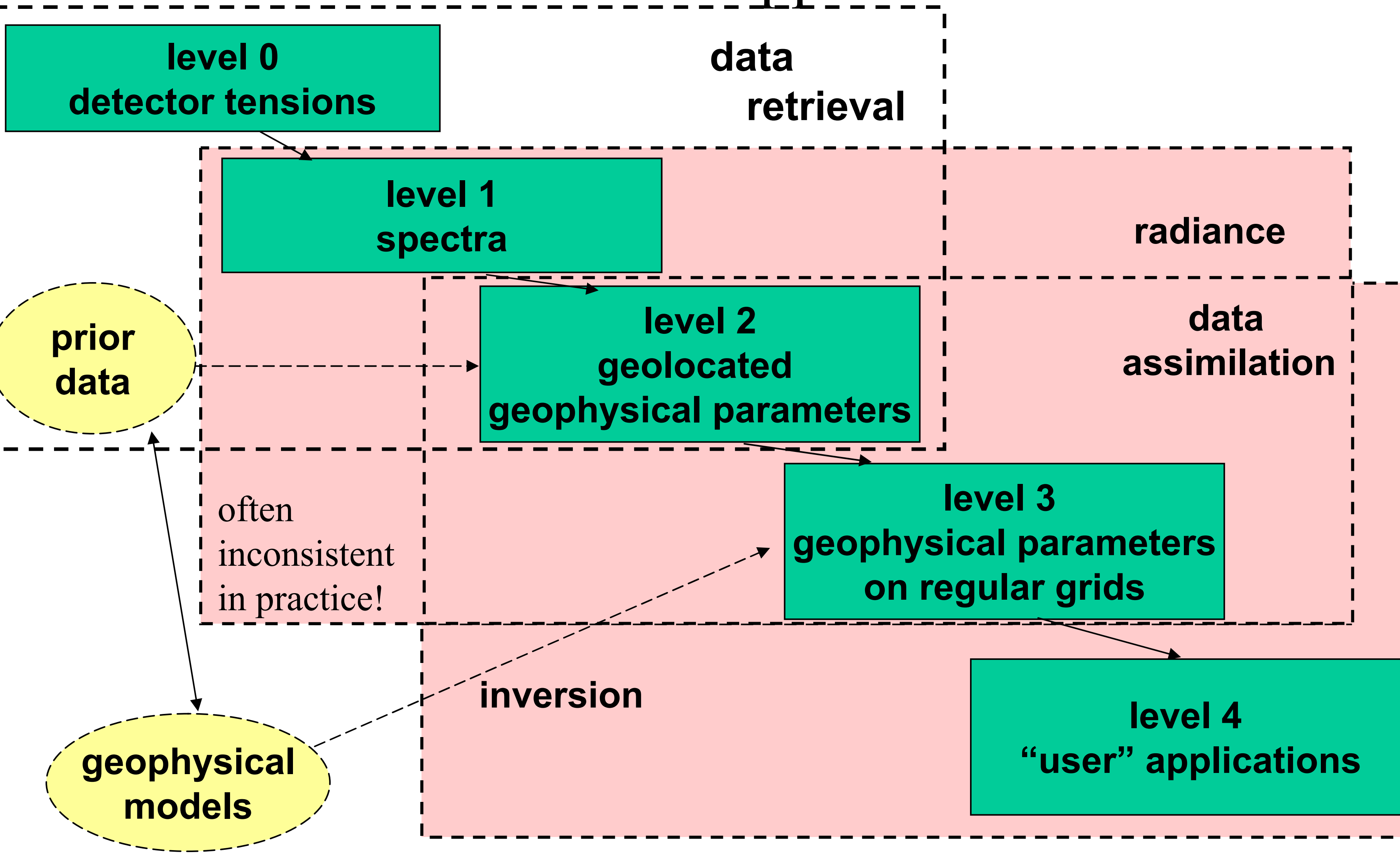
without assimilation



with assimilation



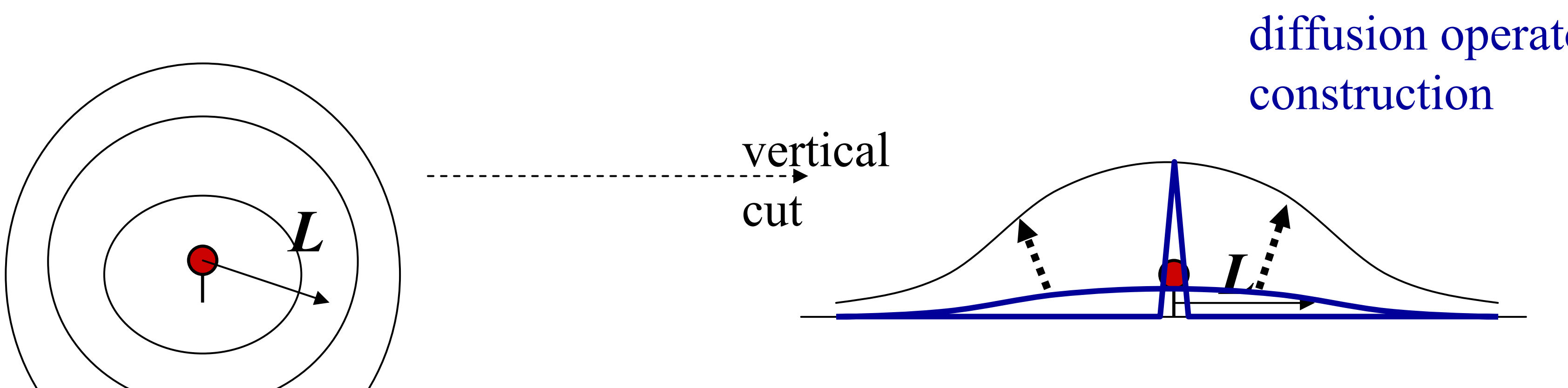
trop. chemical DA in the satellite data application chain



Radius of Influence ((de-)correlation length): Extending the information from an observation location

Textbook:
horizontal influence radius ***L***
around a measurement site,
to be based on a priori
statistical assessments

1D horizontal structure function,
to be stored as a column of the
forecast error covariance matrix

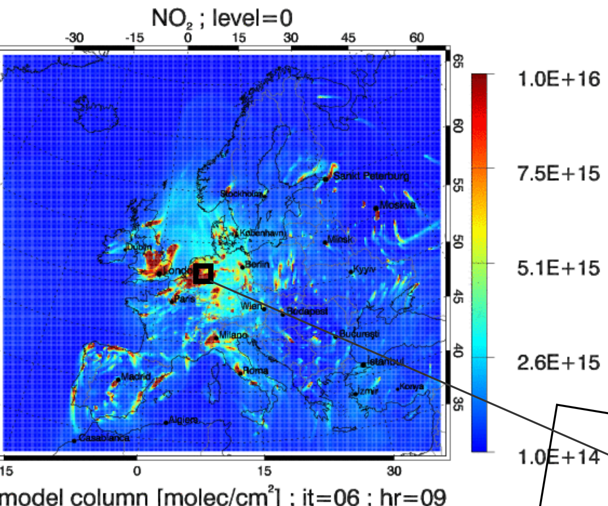


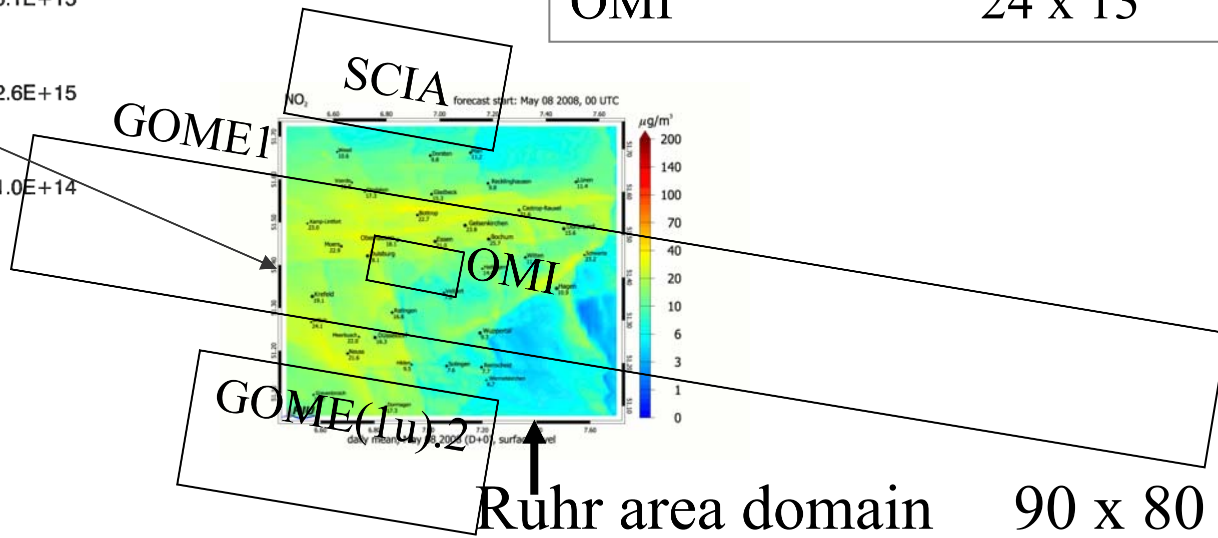
GLOBMODEL case study
NO2 column focussed

- resolution to meet OMI:
  → **15 km** horizontal resolution selected
- attention to forecast error covariance design:
  →spatial correlation exploitation via **inhomogeneous and anisotropic** radii of influence,
- DA method: chemical 4D-var as BLUE, incl emissions, with **externally** provided a prioris:
  → NO2 columns errors from data provider, model error from other case studies, i.e. **no "tuning"** introduced

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 “



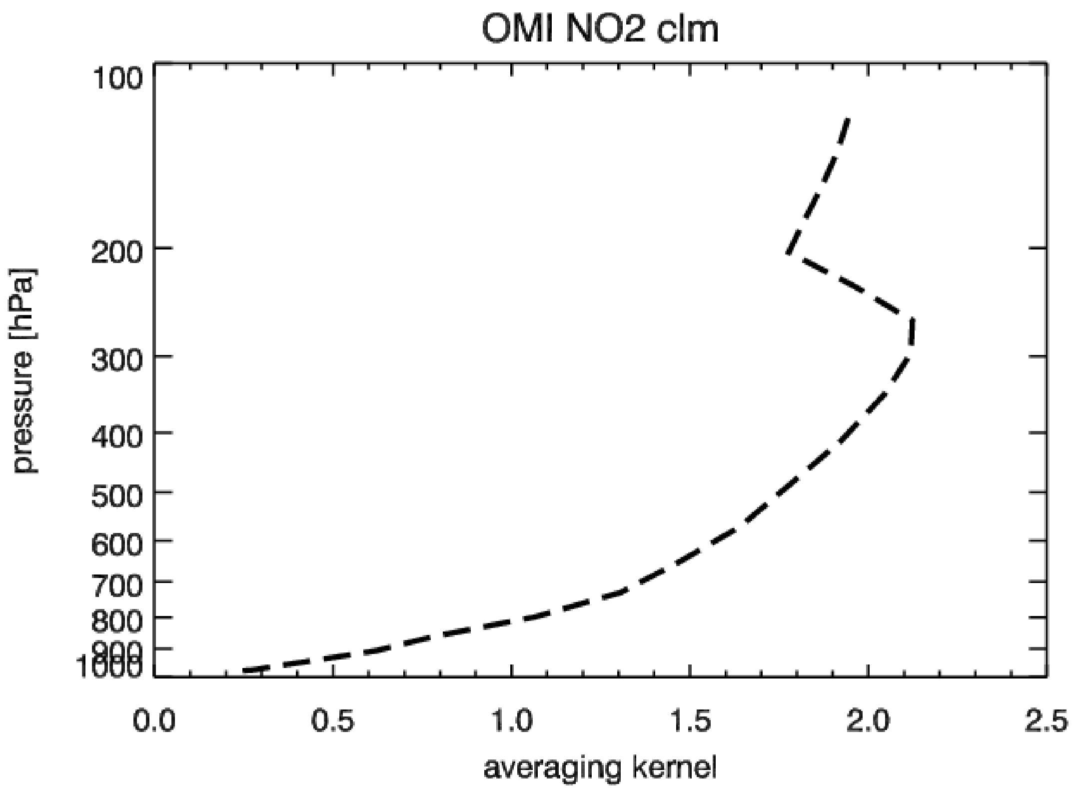


Ruhr area domain 90 x 80 km

1 km resolution

(~12 000 000 inhabitants)

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



model domain mean averaging kernel.

Exploitation of NO2 column averaging kernel information

- shape largely dependent on optical properties of the atmosphere (cioud 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

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 destroying the BLUE property of the assimilation algorithm?

Observation operator $\mathbf{H}$

Formally an integral equation to be solved for vertical NO_2 molecule density
unction x

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

iscretisation

$$y = \sum_{k=1}^{K} h_k x_k$$

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:

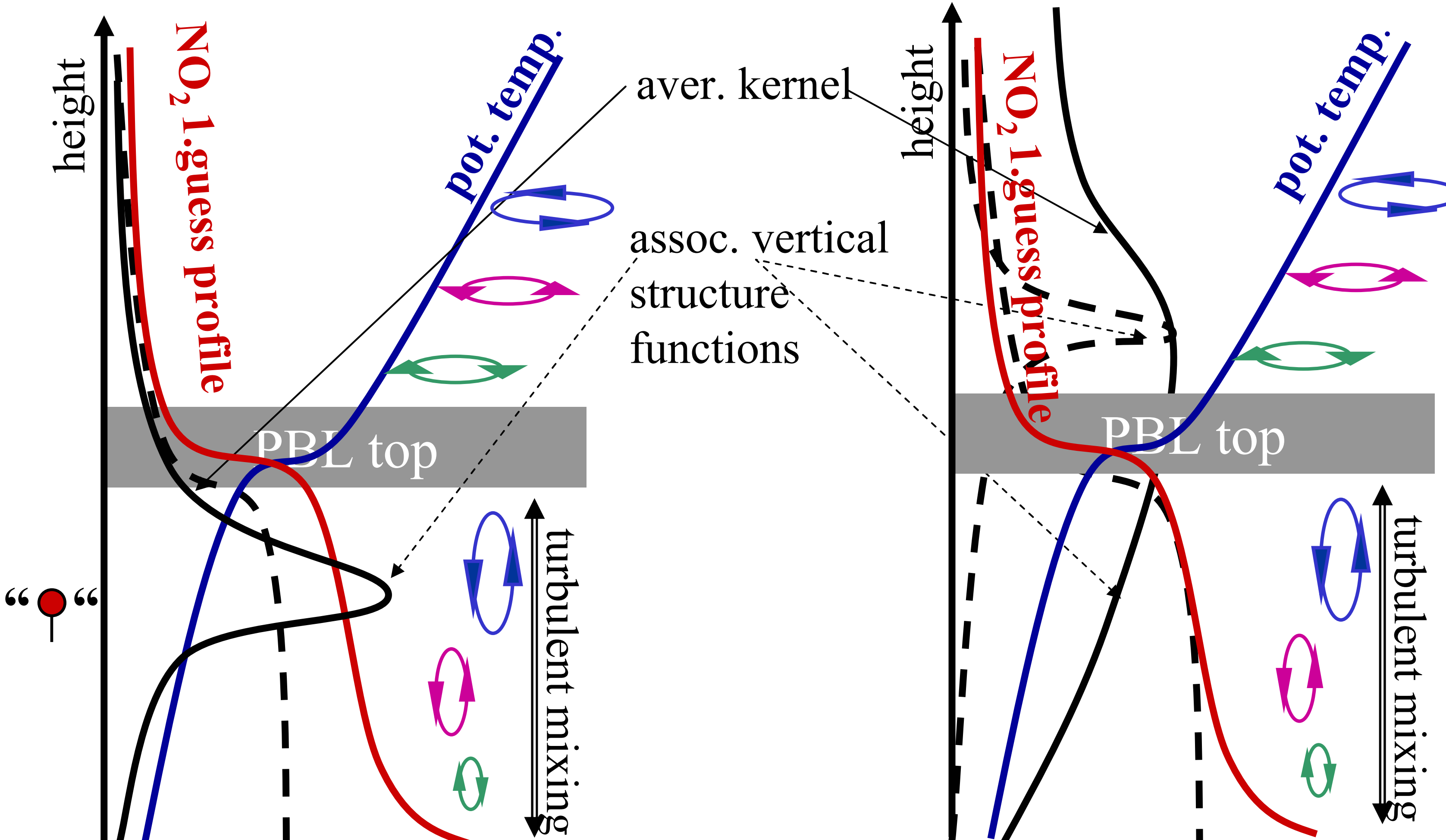
$$\boxed{dx_a = \underbrace{\mathbf{B}\mathbf{h}^T(r+b)^{-1}}\left\{y^0 - H[\mathbf{x}_b]\right\}}$$

adjoint "representer" (oceanographic DA parlance)

→ vertical structure function in B essential!

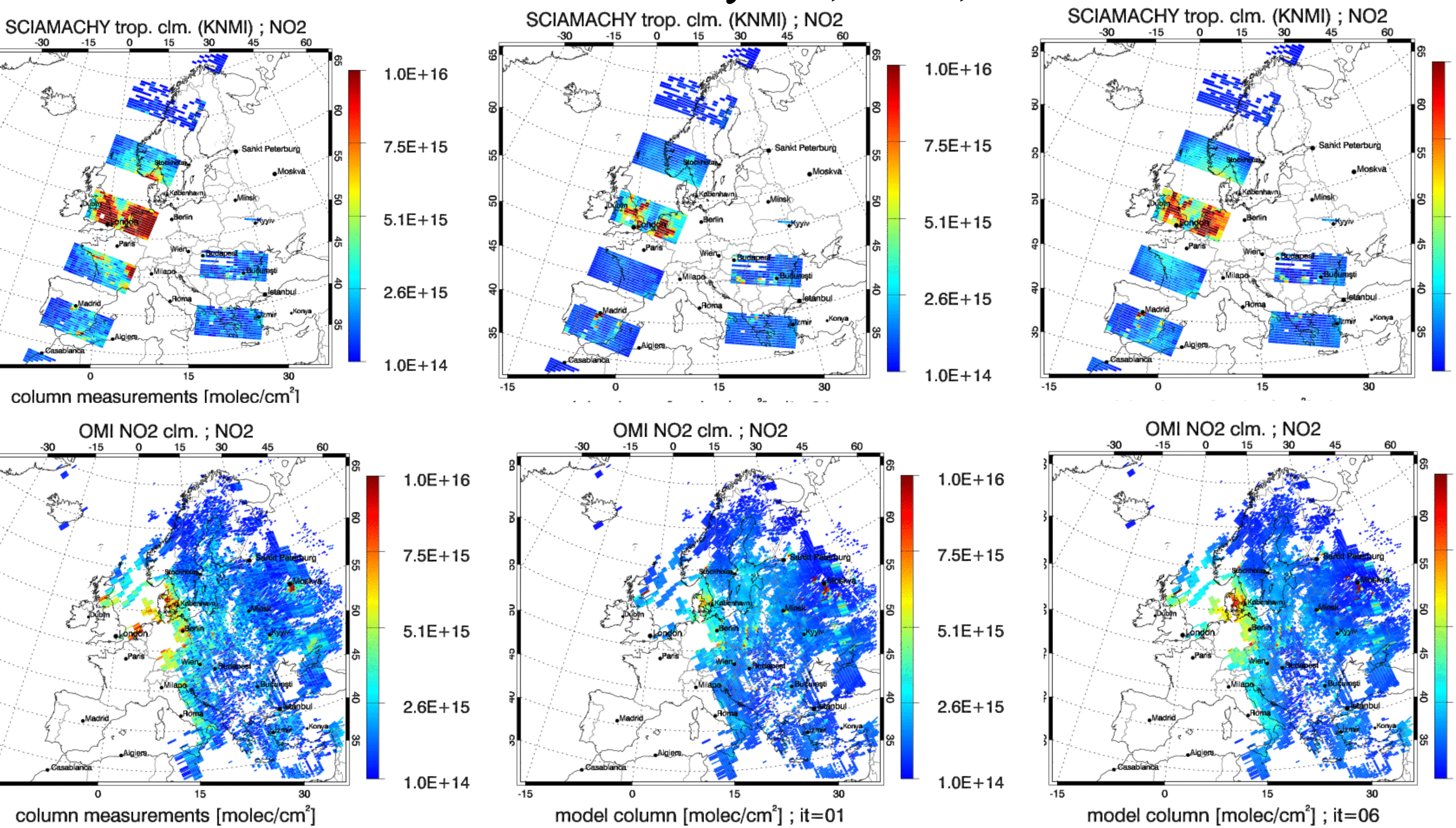
vertical Radius of Influence:
Extending the information from observation location

ideal case real



Comparison of NO2 tropospheric columns

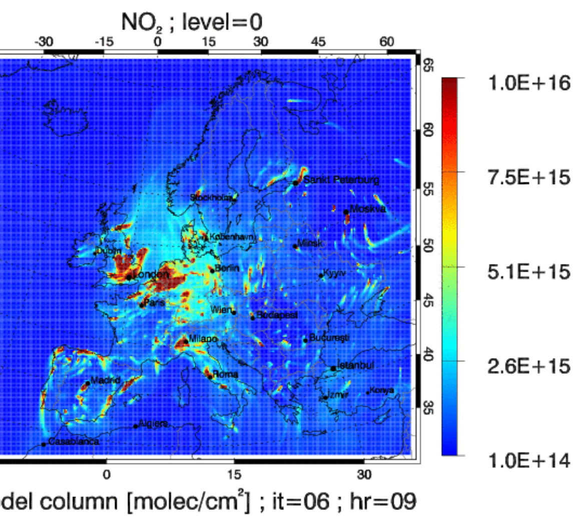
in molecules/cm^2 for July 6th, 2006, 09-12 UTC.

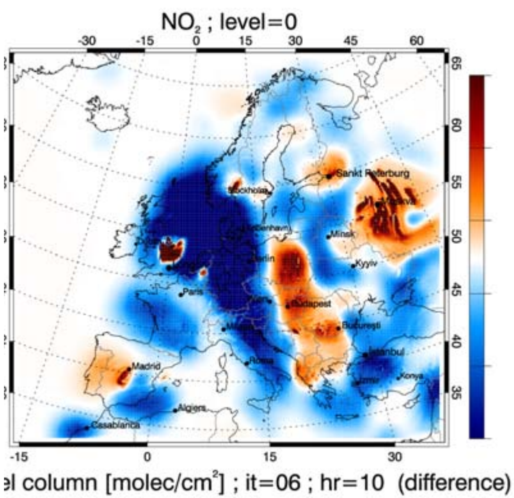


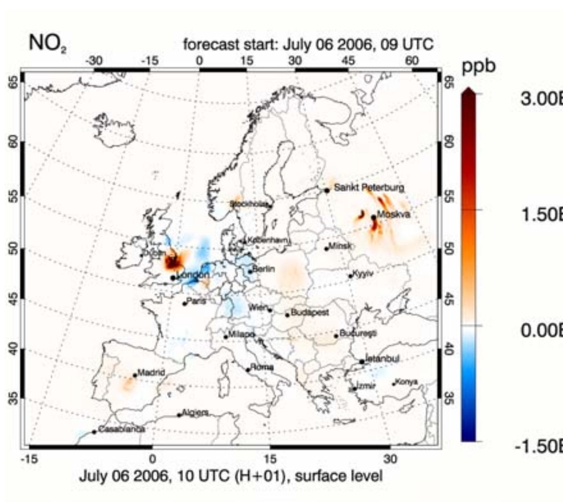
ssimilated values (y) | EURAD forecasted (**Hx**) | column analyses (**H**

Data assimilation result from tropospheric columns for **July 6th, 2006**.

NO2 model columns by OMI and SCIAMACHY assimilation interval 09-12 UTC.





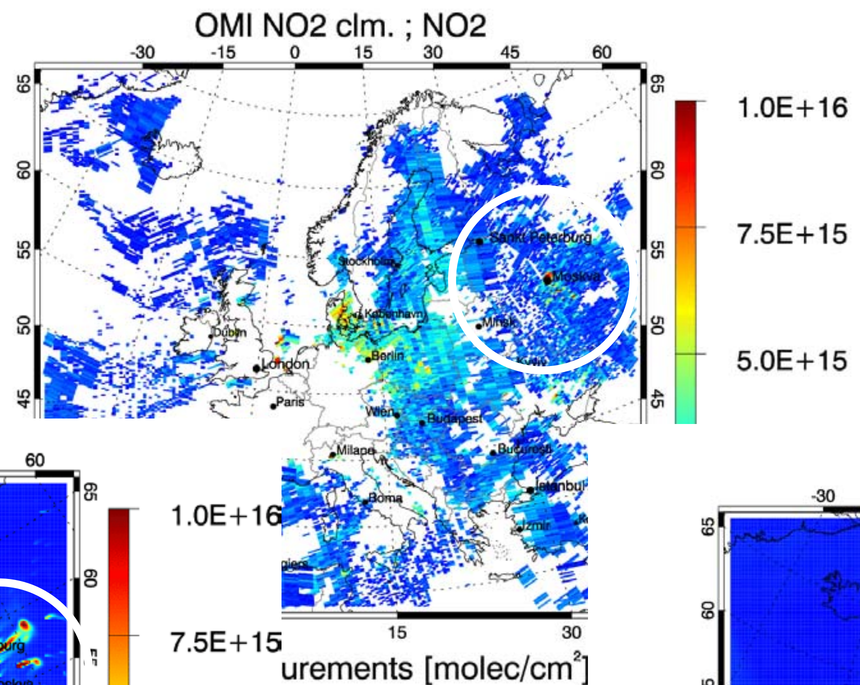


NO2 p

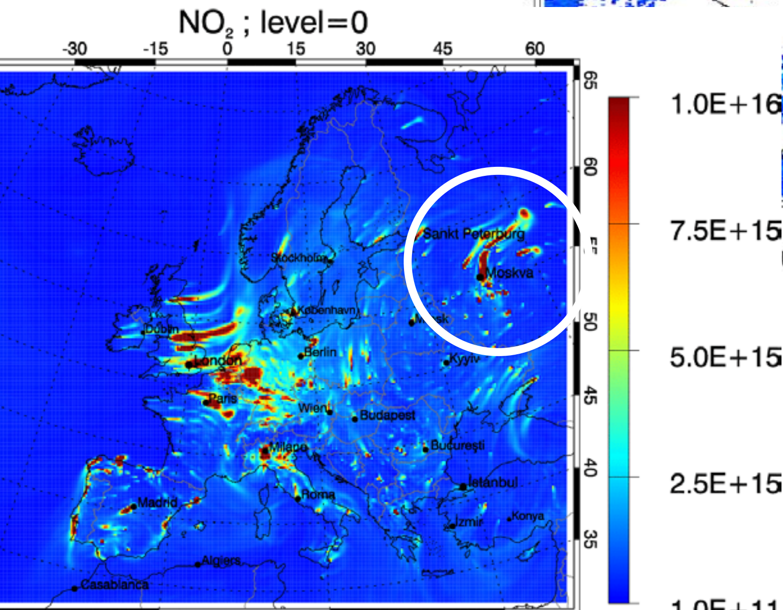
molecules/cm^2

Analysed NO2 colum | Difference field | surface concentration changes

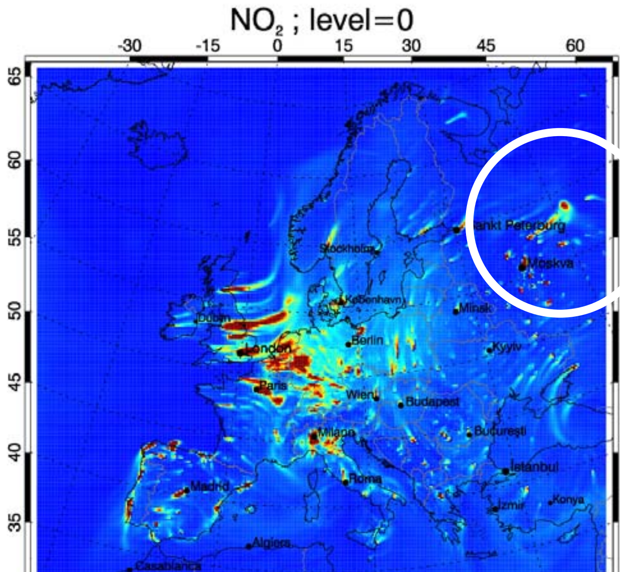
Data assimilation result in terms of tropospheric columns for **July 7th**, 2006. NO2 model columns based on OMI and SCIAMACHY assimilation within the assimilation interval, 09-12 UTC.



ure forecast



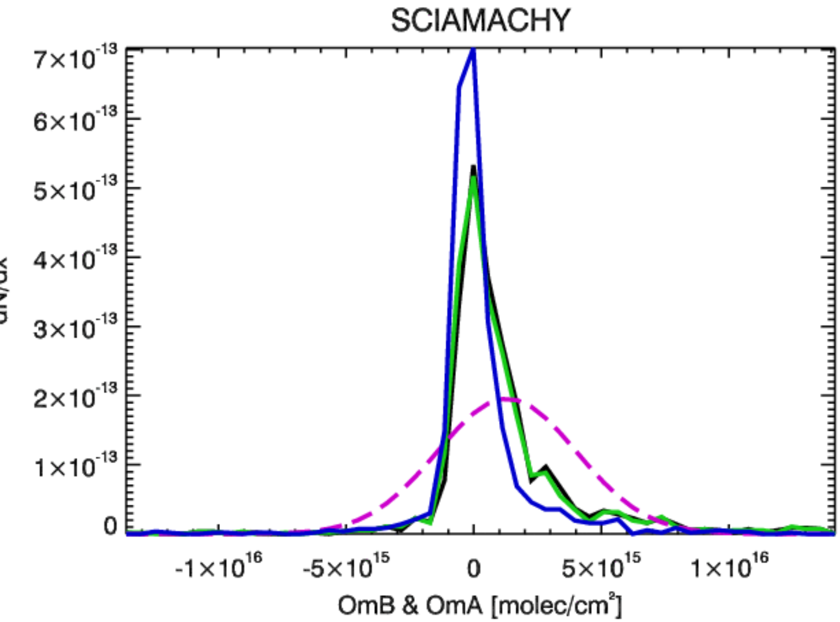
assimilation based forecast

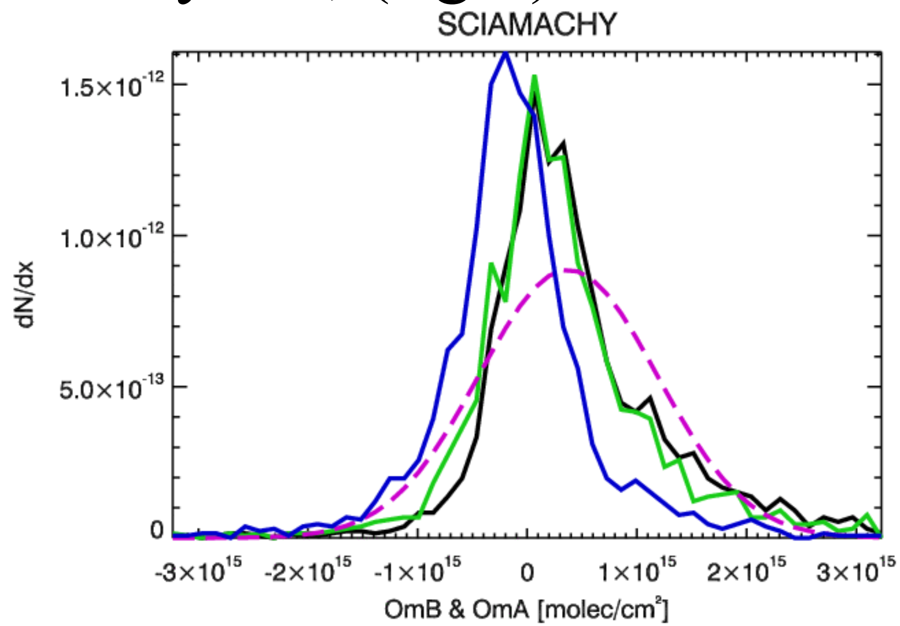


SCIAMACHY

$O_{SCIA}mX_{OMI}$ probability density functions

for July 6th (left), and July 8th, (right).





Control run (OmC) (no data assimilation at all,) black bold line,
assimilation based forecasted values (OmF) green bold line,
analyses (OmA) blue bold line.
For comparison: Gaussian fit to OmF pdf by
mean and standard deviation given by broken purple line.

Conclusions: Did we mature?

- data assimilation to be extended toward <u>general inversion</u>
- emission rate estimation feasible, other parameters to be confirmed: <u>improved preconditioning mandatory</u>
- covariance matrix problem: how to implement <u>multivariate optimisation parameters</u>
- tropospheric column information <u>still to be optimized</u> for successful surface observation validation