Workshop on parameter estimation and inverse modelling for atmospheric composition

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for atmospheric composition
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Estimating emission rates of reacting constituents by variational inversion

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Contents

Data assimilation and Inverse Modelling: What makes the difference?

- **Data assimilation**: estimate
  - transient parameters:
  - prognostic parameters, state variables of prognostic systems
  - **for reactive chemistry: <u>initial values</u>**
- **Inverse Modelling**: estimate
  - intransient system control parameters,
  - forcing parameters ,
  - diagnostic parameters
  - **for reactive chemistry: <u>emission rates, (deposition velocities)</u>**

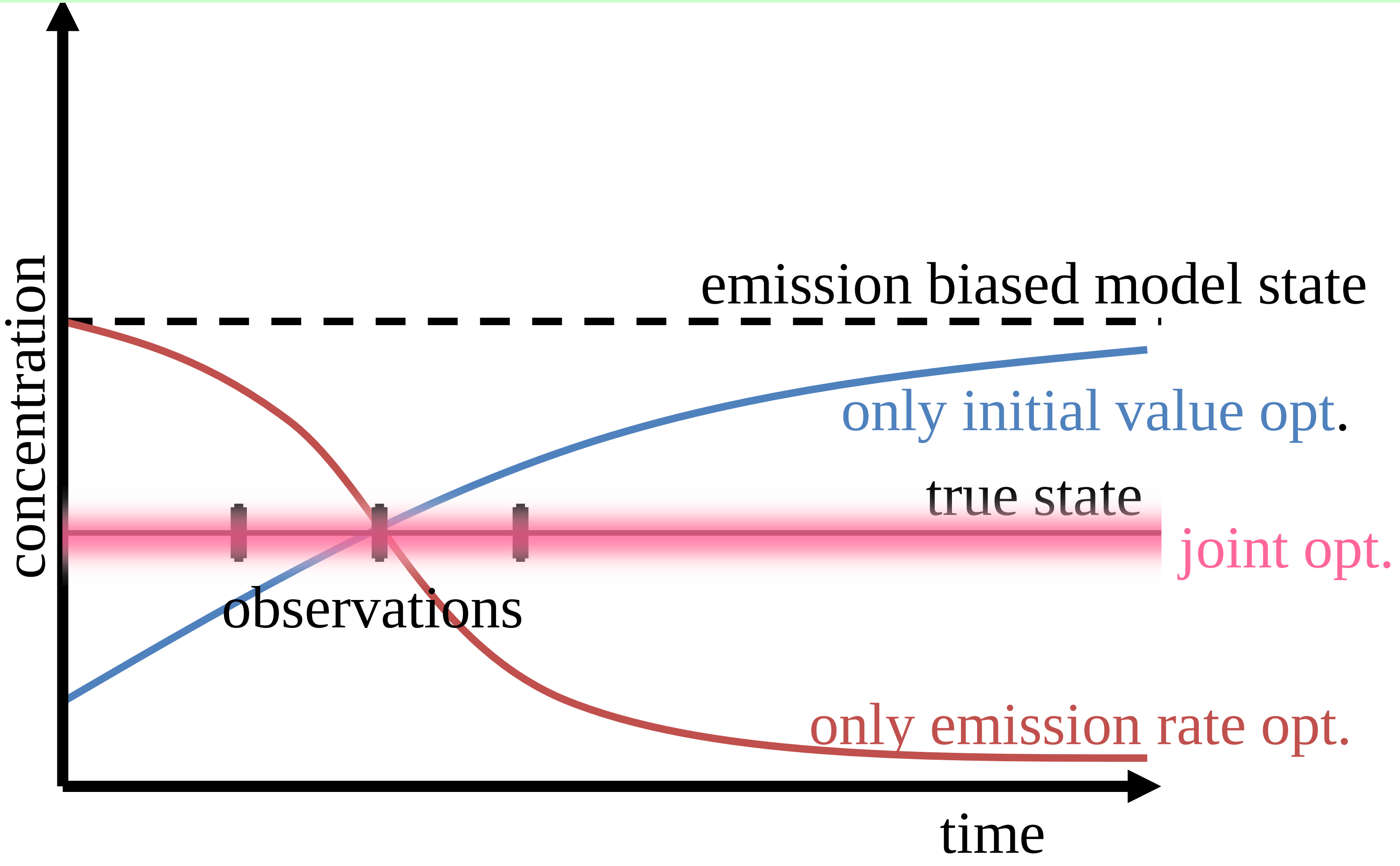
➔time and spatial scales carefully to be considererd, if combined

Problem formulation (increasingly ill-posed)

- which parameter sets are to be optimized at all, and are the observations available?
- poor representativity of observations
- often only integral measurements available (NB: PM_x, $NO_{x/y}$, column soundings from satellites)
- reactive chemistry, and its observabiliy (with poorly observed emitted compounds)

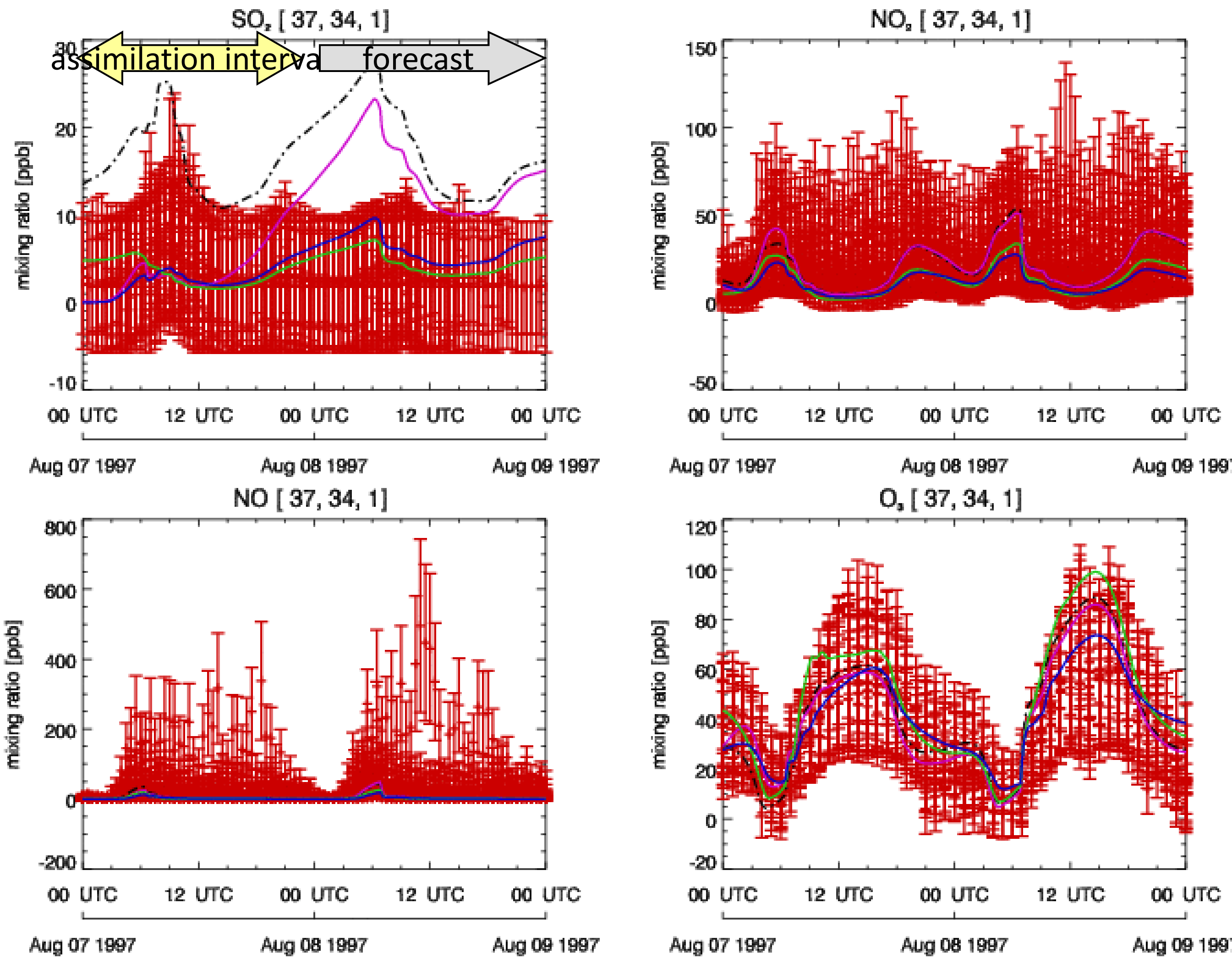
SPECIFIC PROBLEMS

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



The representativity problem (1): non-Gaussian distribution example: Rhine-Main area box: (Frankfurt-Mainz)

9.-10. August 1997



+ observations
no optimisation

initial value opt.

emis. rate opt.

joint emis +
ini val opt.

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:

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

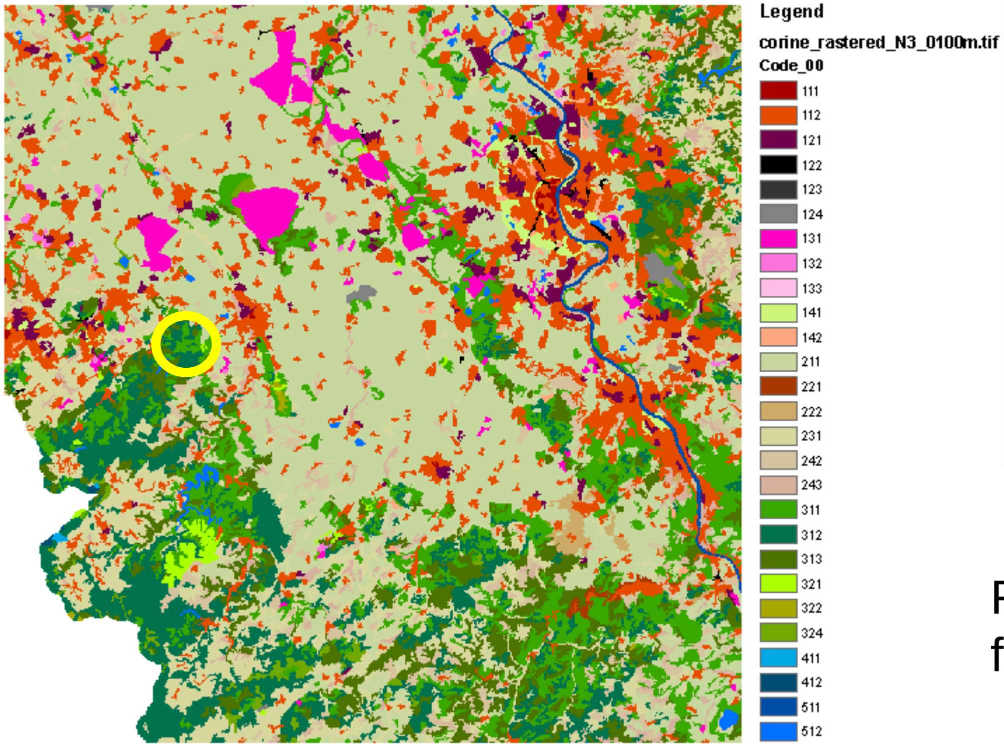
- Gradient:

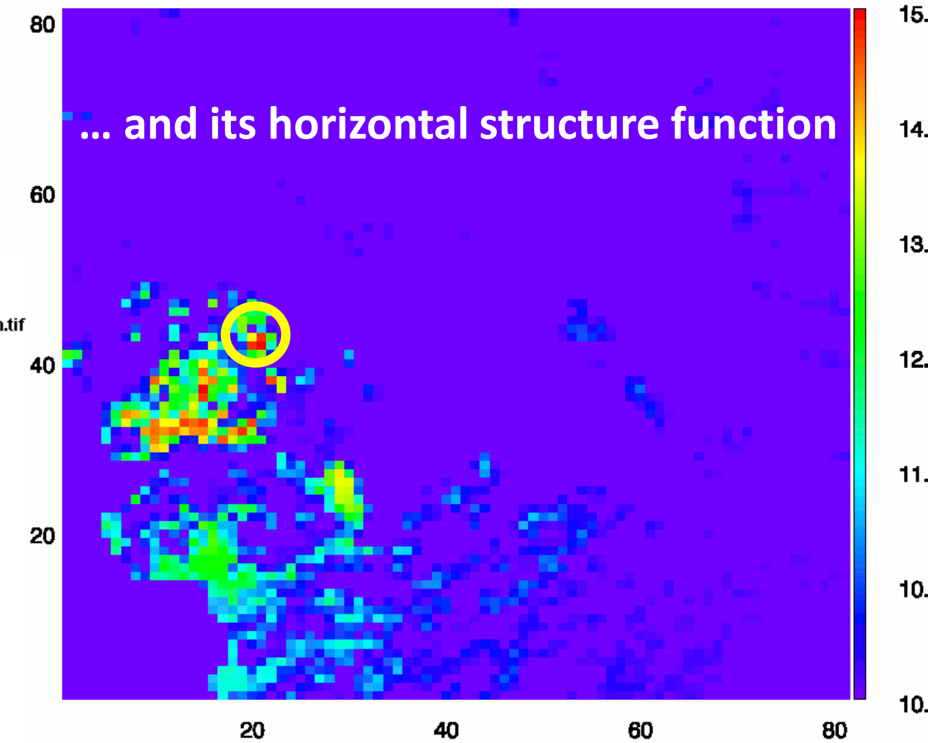
$$\nabla_{\boldsymbol{v}}J = \nabla_{\boldsymbol{v}}J_{IV} + \nabla_{\boldsymbol{v}}J_O = \boldsymbol{v} + \mathbf{B}^{\mathrm{T}/2}\nabla_{\delta\boldsymbol{v}}J_O$$
$$\nabla_{\boldsymbol{w}}J = \nabla_{\boldsymbol{w}}J_{EF} + \nabla_{\boldsymbol{w}}J_O = \boldsymbol{w} + \mathbf{K}^{\mathrm{T}/2}\nabla_{\delta\boldsymbol{w}}J_O.$$

The representativity problem (2):

Tesselated terrain and associated covariances

High resolution CORINE land use of the Cologne Aix-la-Chapelle area with a forest harboured station

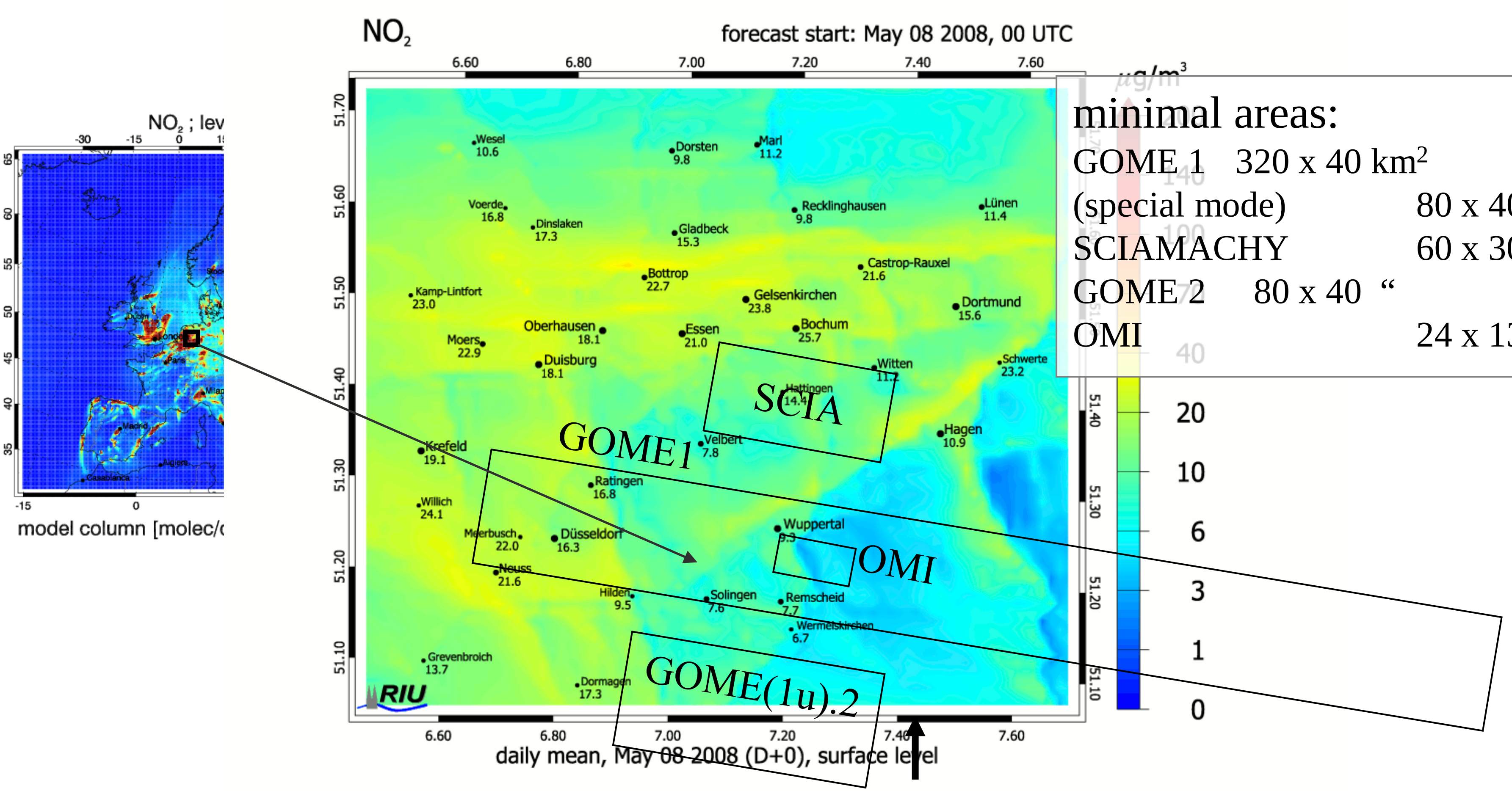




For high resolution, correlation functions need to bridge gaps

Integral information - Satellite data:

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 90 km^2 x 80 km^2

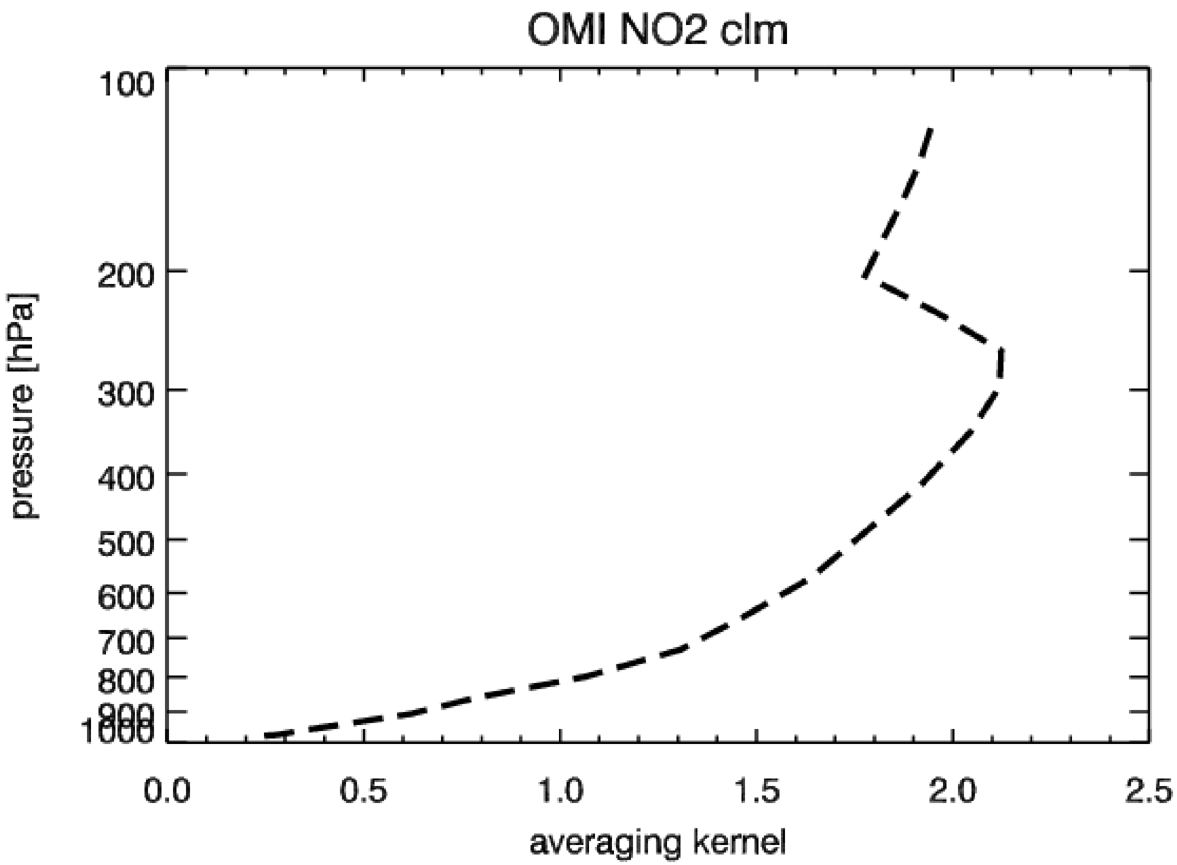
(~12 000 000 inhabitants)

Integral information –

Do NO2 columns profice useful information?

Average OMI averaging kernel profile

over model domain for July 9th, 2006



model domain mean averaging kernel.

Integral information –
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

Integral information – Observation operator **H**

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

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

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

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

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

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

For scalar column retrieval:

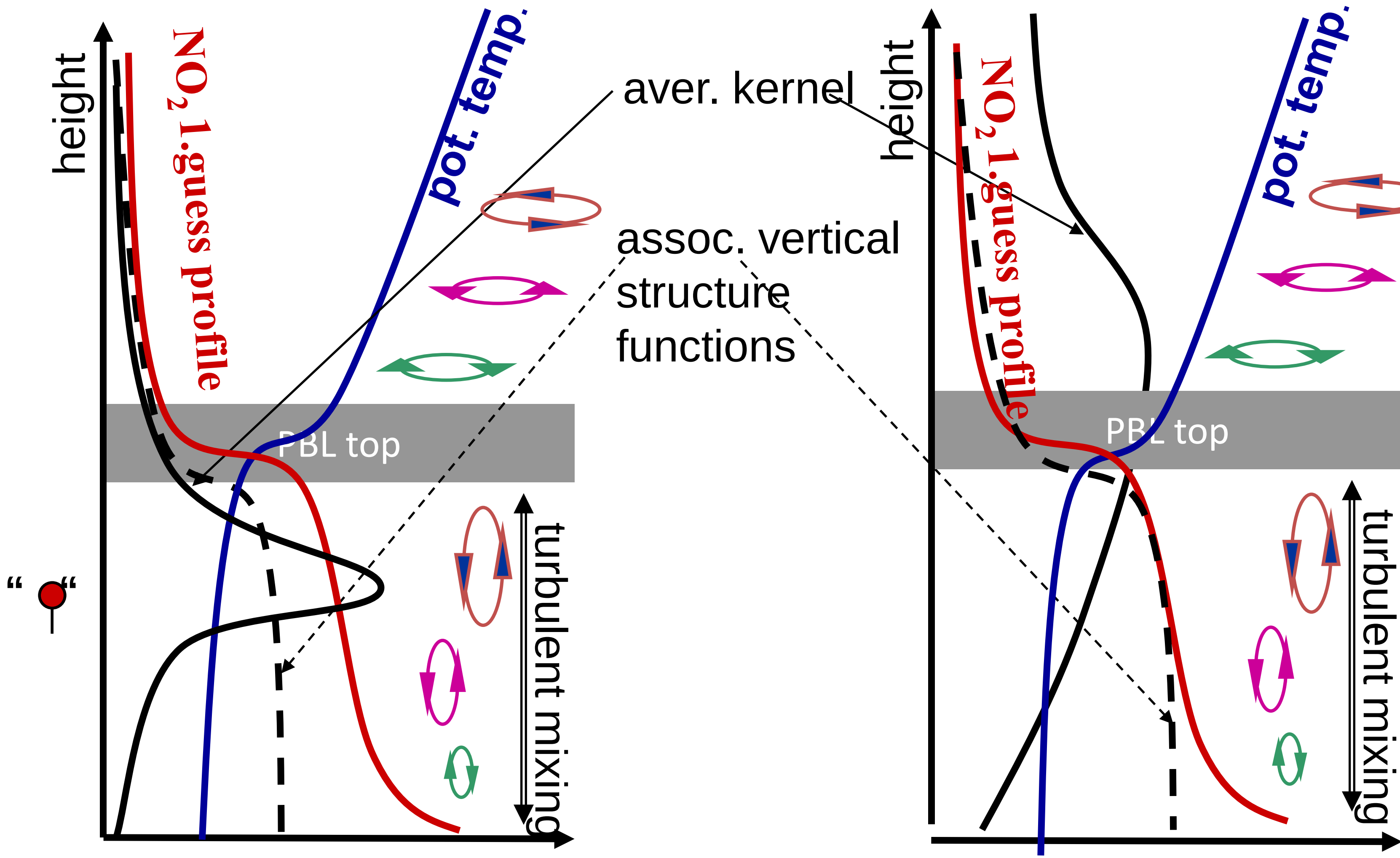
$$d\mathbf{x}_a^c = \underbrace{\mathbf{B}\mathbf{h}^T(r+b)^{-1}}\{y^0 - h[\mathbf{x}_b]\}$$

adjoint representer

→ vertical structure function in **B** essential!

Vertical structure function:
Extending the information from observation location by vertical exchange of polutants and information

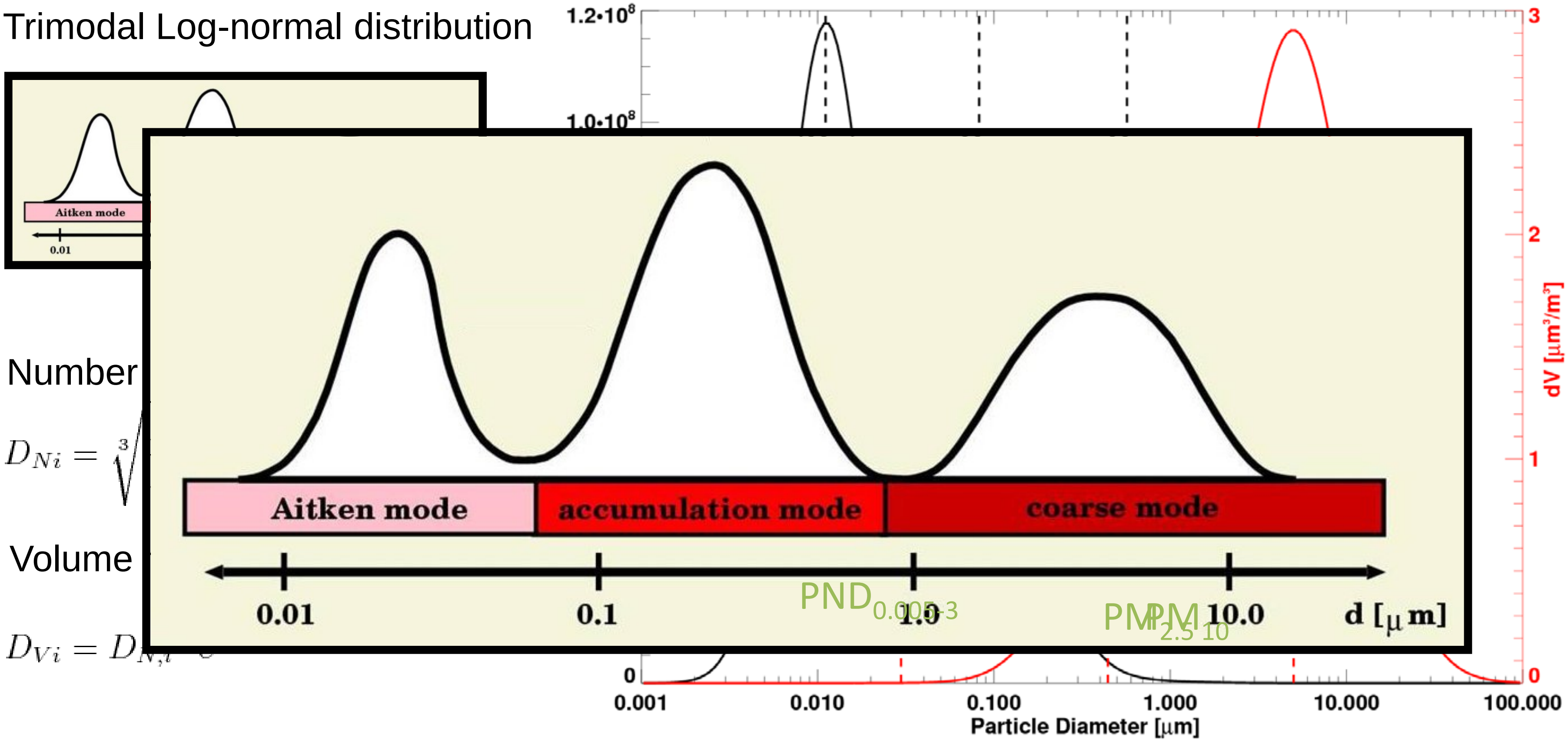
ideal ***case*** ***real***



Integral information –
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

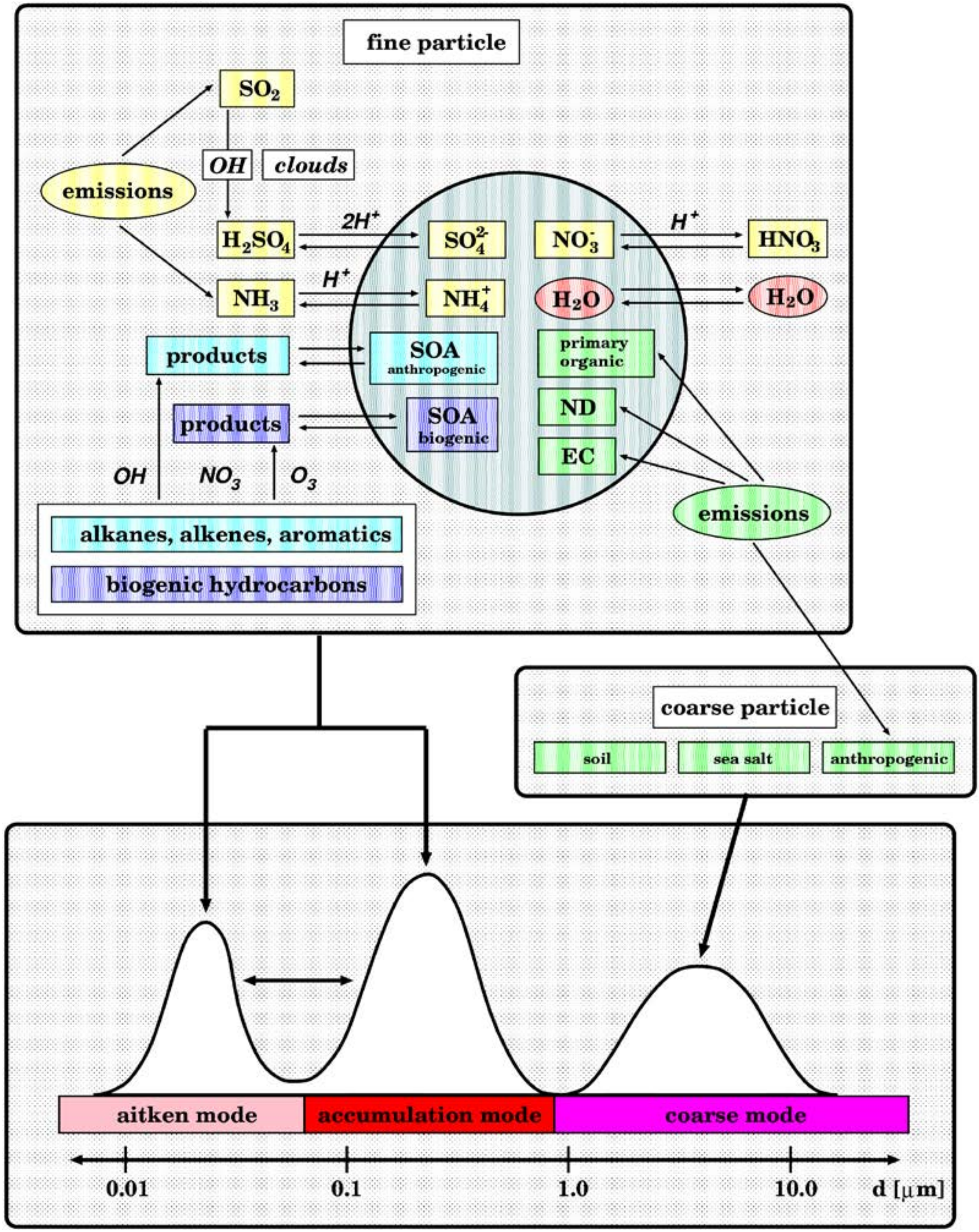
Integral information -

Example:
Aerosol Chemistry in
MADE

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

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

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



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

MERIS MODIS
AATSR+SCIAMACHY,...

SYNAER retrieval algorithm

Species Mapping

EURAD-IM [µg/m³]	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

Further significant problems, not considered here

- Meteorological driver model with boundary layer, convection, and precipitation
- chemistry model errors
- deposition velocities
- ….

ISSUES ON OBSERVABILITY

Observability
ad a simple forcing model

Consider two one-way coupled linear models:
$A(t_i, t_0)$ atmospheric model,
$E(t_i, t_0)$ emission model, and
C the coupling between both.

$$\begin{pmatrix} H(t_i), & 0 \end{pmatrix} \begin{pmatrix} x(t_i) \\ e(t_i) \end{pmatrix} = \begin{pmatrix} H(t_i, t_0), & 0 \end{pmatrix} \underbrace{\begin{pmatrix} A(t_i, t_0), & C \\ 0, & E(t_i, t_0) \end{pmatrix}}_{:=M(t_i, t_0)} \begin{pmatrix} x(t_0) \\ e(t_0) \end{pmatrix}$$

$M(t_i, t_0)$ model integration from time t_0 to t_i
$H(t_i)$ observation operator at time t_i
$x(t)$ atmospheric model state,
$e(t)$ emission parameter.

Observability

$H(t_i)$ observation operator at time t_i

$$O_v := \begin{bmatrix} H(t_0) \\ H(t_1)M(t_1, t_0) \\ H(t_2)M(t_2, t_0) \\ \vdots \\ H(t_v)M(t_v, t_0) \end{bmatrix}$$

Consider rank of matrix, Singular Vectors and -Values of

$$O_v^T O_v$$

to be above some noise level.

How can we identify the degree of observability?

Given CTM (here RACM and EURAD-IM) acting as tan.-lin. model operator $\mathcal{L}$:

$$\delta\mathbf{c}(t_F) = \mathcal{L}_{t_I,t_F}\,\delta\mathbf{c}(t_I), \quad \mathcal{L}_{t_I,t_F} = \left.\frac{\partial \mathcal{M}_{t_I,t_F}}{\partial \mathbf{c}}\right|_{\mathbf{c}(t_I)}$$

1. Berliner et al., (1998) Statistical design:
"Minimize" the analysis error covariance matrix **A** (say, via trace):

$$\min_{\mathbf{H}} \mathbf{A} = \mathbf{B} - \underbrace{\mathbf{B}\mathbf{H}^T(\mathbf{H}\mathbf{B}\mathbf{H}^T + \mathbf{R})^{-1}\mathbf{H}\mathbf{B}}_{\text{to be maximized by } \mathbf{H}}$$

For this find maximal eigenvectors as observation operators **H,** which configure observations.

$$\mathcal{L}_{t_I,t_F}\mathbf{B}\mathcal{L}^T_{t_I,t_F}\mathbf{H}^T = \lambda\mathbf{H}^T$$

2. Palmer (1995) Singular vector analysis:
Observe maximal SV configuration:

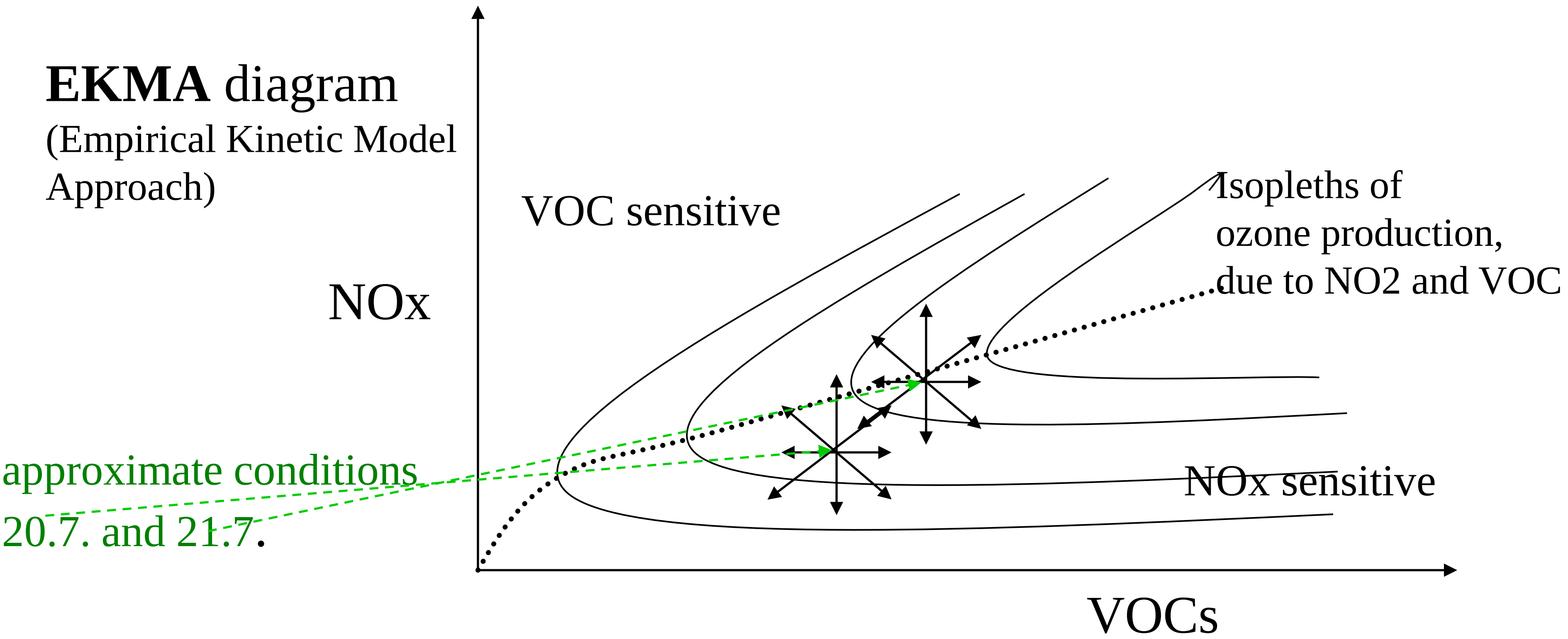
$$\max_{\delta\mathbf{c}(t_I)} \frac{\|\delta\mathbf{c}(t_F)\|^2_{\mathbf{B}}}{\|\delta\mathbf{c}(t_I)\|^2_{\mathbf{B}}} = \max_{\delta\mathbf{c}(t_I)} \frac{\delta\mathbf{c}(t_I)^T\mathcal{L}^T_{t_I,t_F}\mathbf{B}\,\mathcal{L}_{t_I,t_F}\delta\mathbf{c}(t_I)}{\delta\mathbf{c}(t_I)^T\mathbf{B}\,\delta\mathbf{c}(t_I)},$$

A prototype non-linearity example:

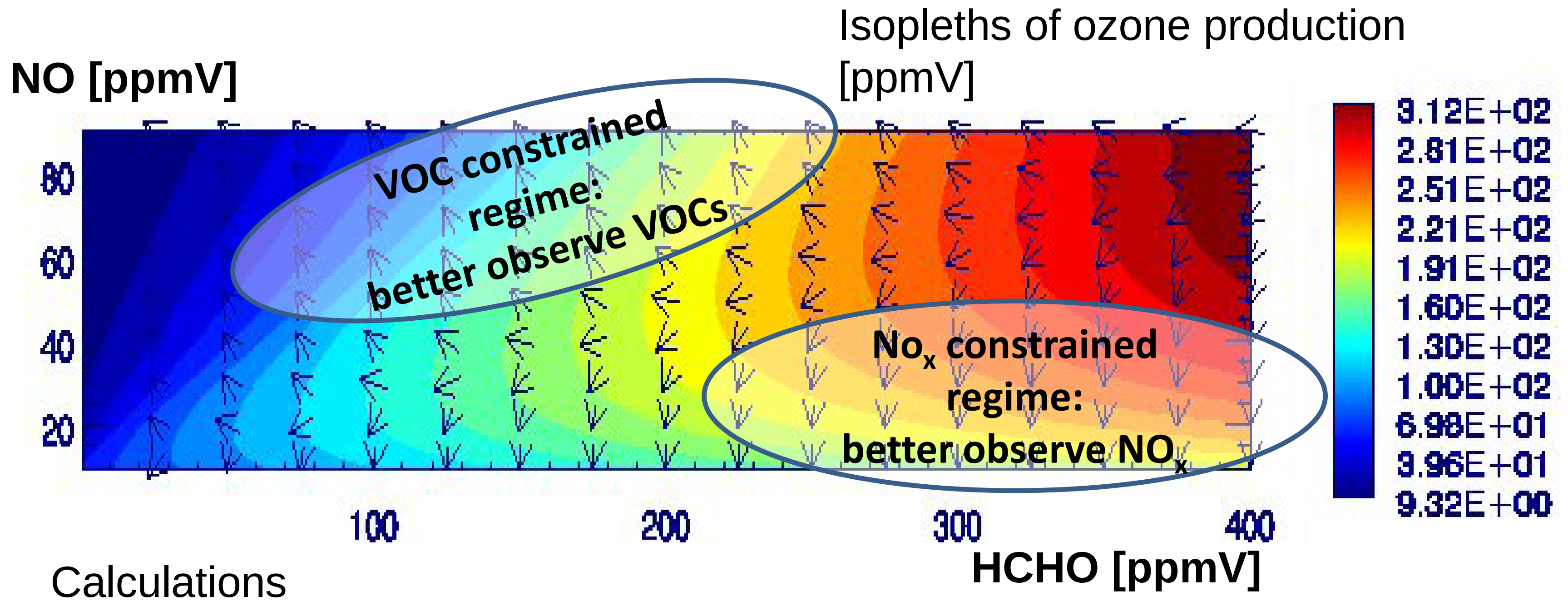
Empirical Kinetic Model Approach scheme

- Nitrogene oxides and numerous hydrocarbons act highly nonlinearly as precursors of ozone.
- Chemical conditions are either controlled by NOx or VOC deficit, delineating the "chemical regime".
- Both 4D-var and Kalman filter should start with the proper chemical regime.

EKMA diagram
(Empirical Kinetic Model Approach)



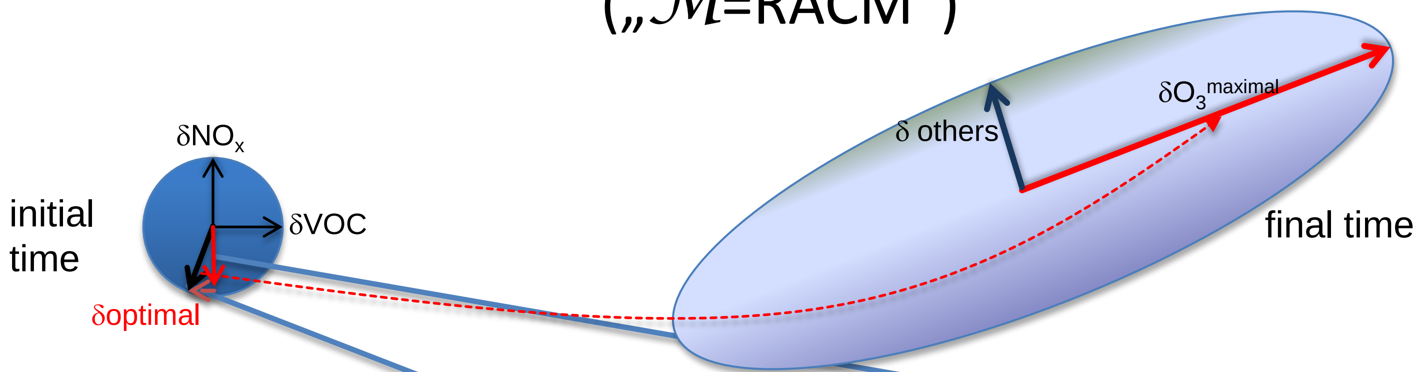
Is NO_x the key to ozone production?
And consequently, its observationthe key to better forecast?



Calculations

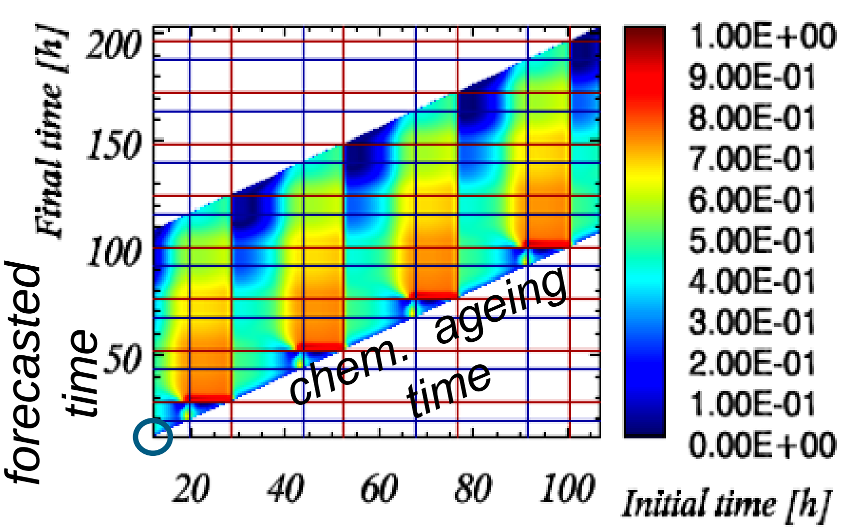
- ✓ within a fixed time span
- ✓ initial conventrations of NO / HCHO were varied
- ✓ change of final concentration is given by colour
- ✓gradients (SVs) of maximyl ozone production given by arrows

Basic 0-D Regional Atm. Chemistry Mechanism („$\mathcal{M}$=RACM“)

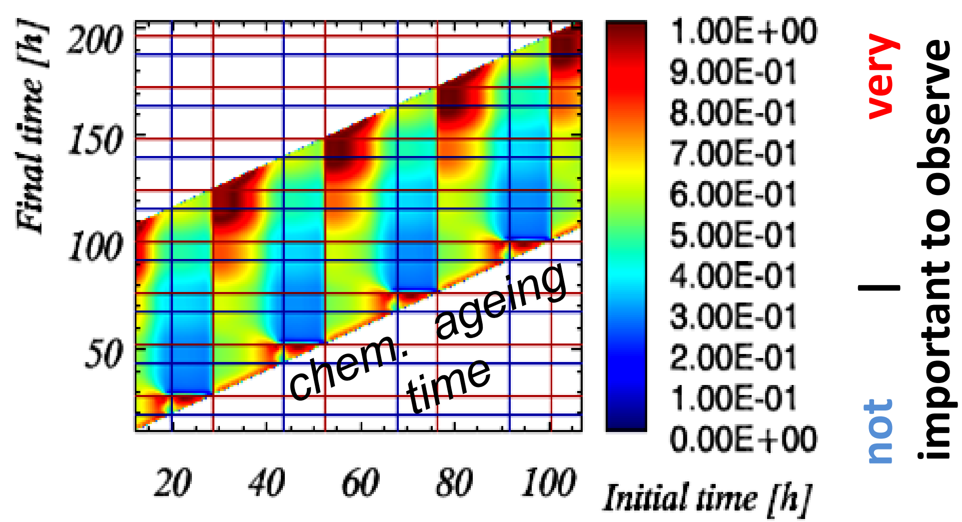


- **Optimal perturbations (Singular Vectors) for** scenario MARINE

1st Grouped Singular Vectors (δVOC)



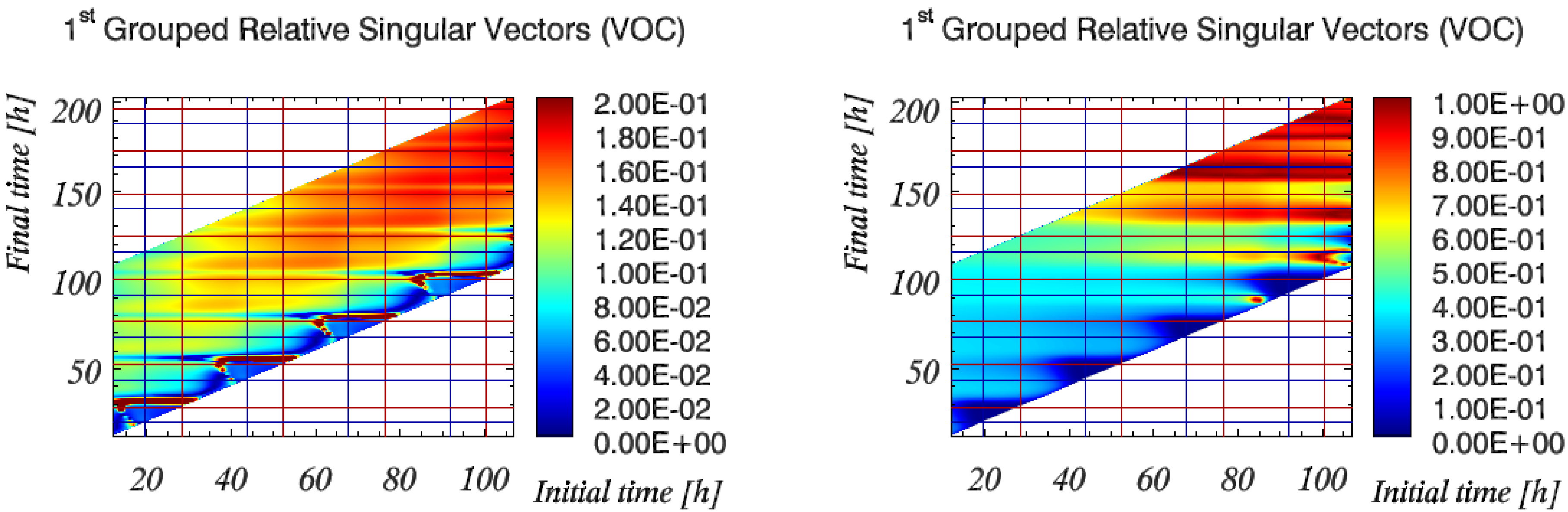
1st Grouped Singular Vectors (δNO$_x$)



sunrise sunset

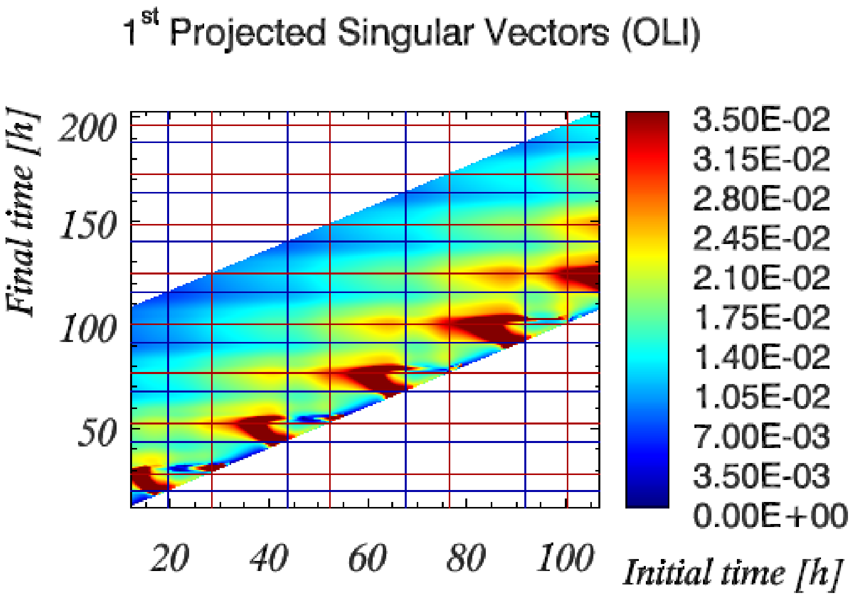
Goris and Elbern, ACPD, 2012

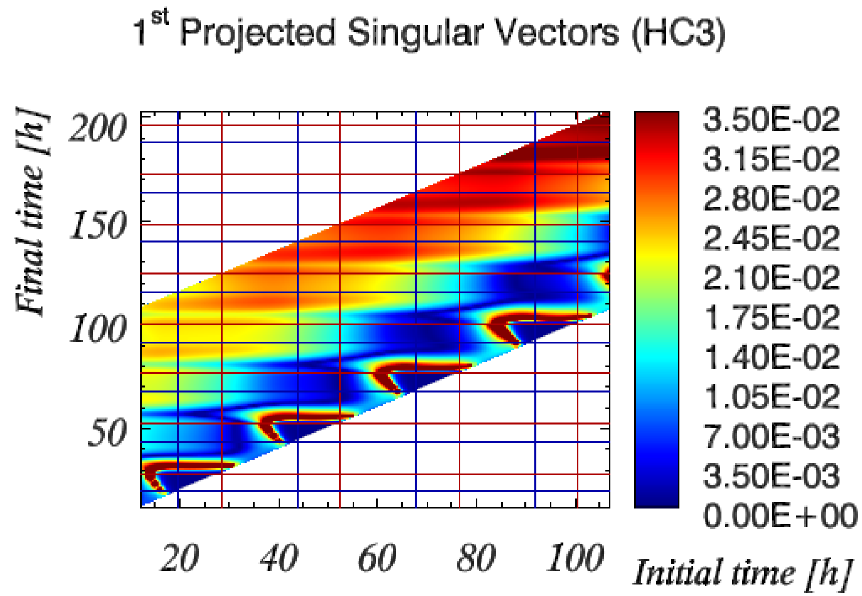
When is the chemical regime sensitive? Leading **emission** singular vectors for VOCs (vs Nox)



TSVD of the optimal grouped relative singular vectors with respect to emission uncertainties for scenarios **(a)** PLUME and **(b)** URBAN.
Both panels depict VOC-sections of the grouped relative singular vectors.

When is the chemical regime sensitive? Leading **emission** singular vectors for olefines (left) and lower alkanes (vs Nox)



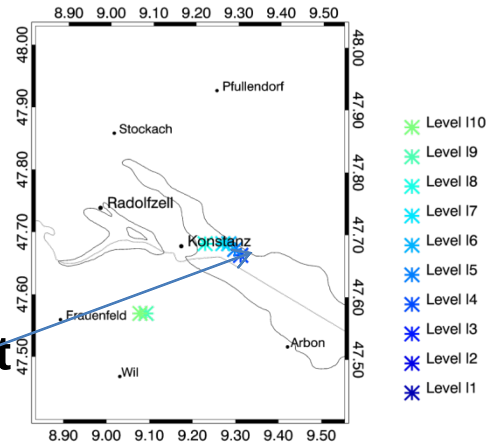


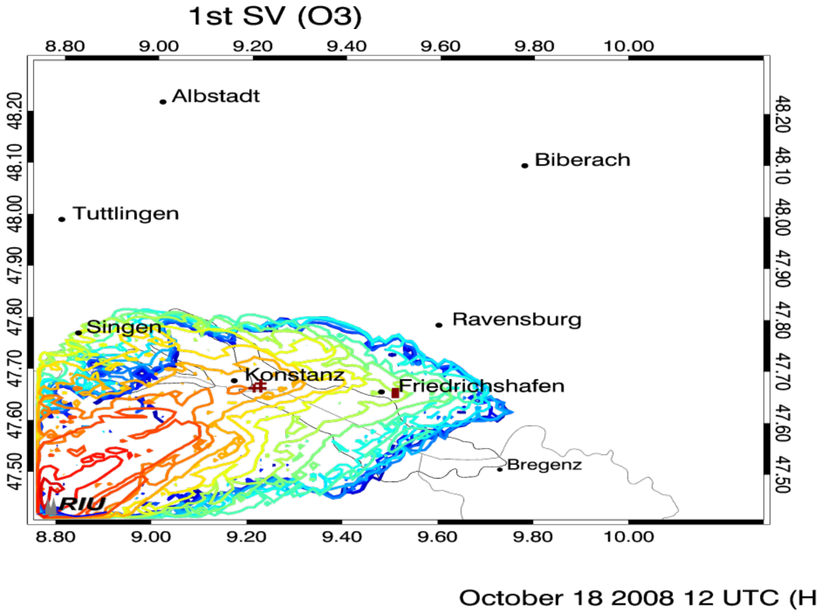
3D-targeted observations for flight missions

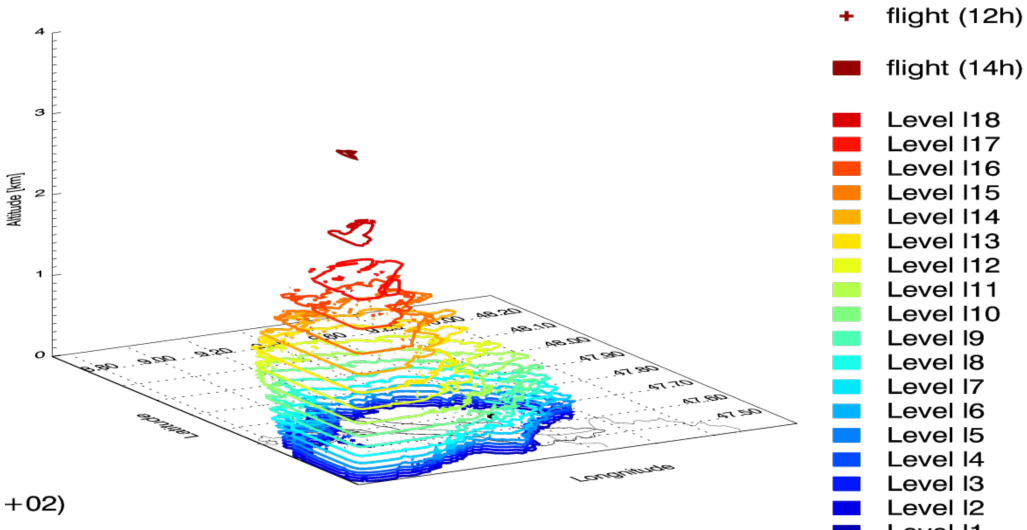
***Given the need for a forecast** at some final time t_F of a mission flight, **what and where should be observed** at the beginning t_I?*

Airship cruising area Lake Constance

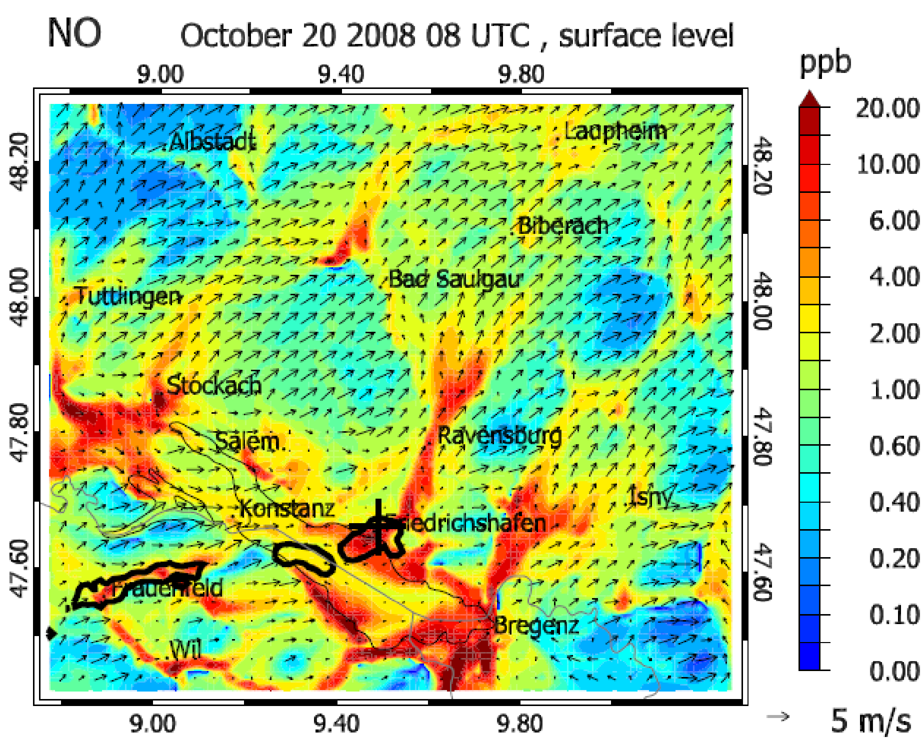
Optimal O_3 forecast seeked for Friedrichshafen airport

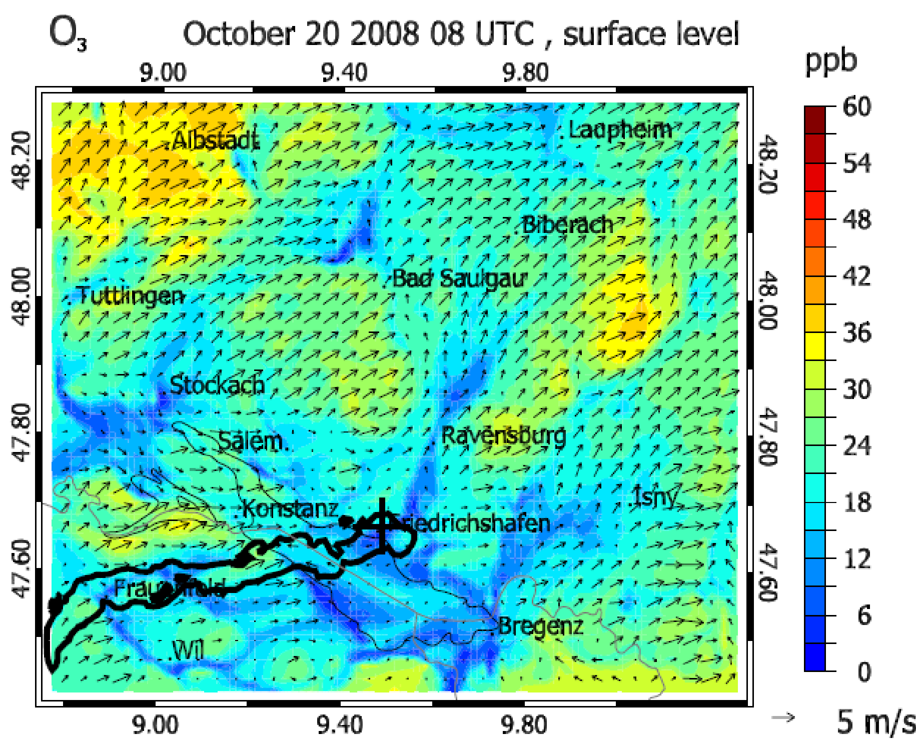






Example observation targeting: SV optimal placement of observation sites





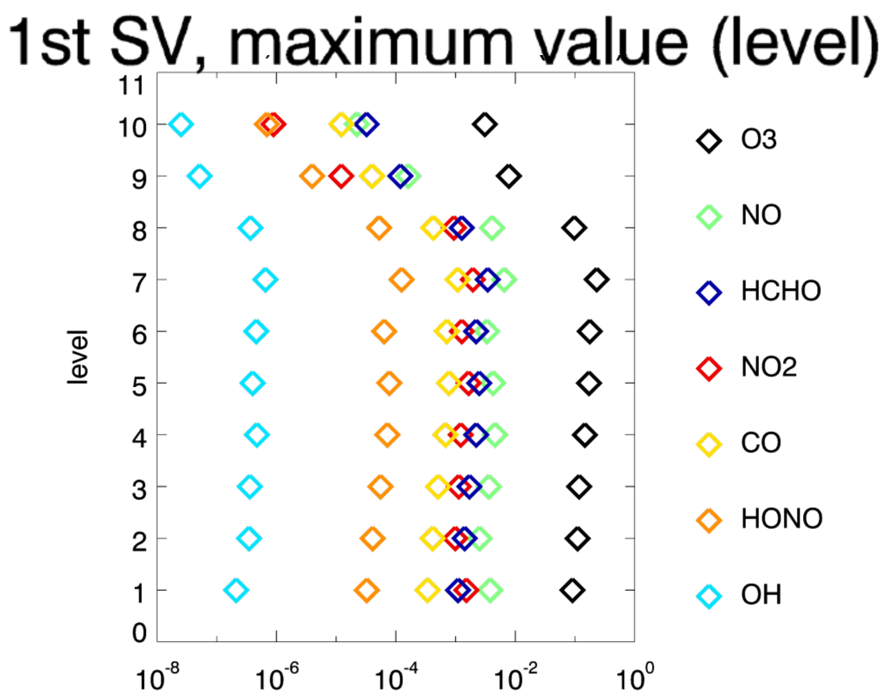
Initial concentrations and optimal horizontal placement of NO (left) and O3 (right) at surface level . Isopleths of the optimal horizontal placement are indicated with black lines.

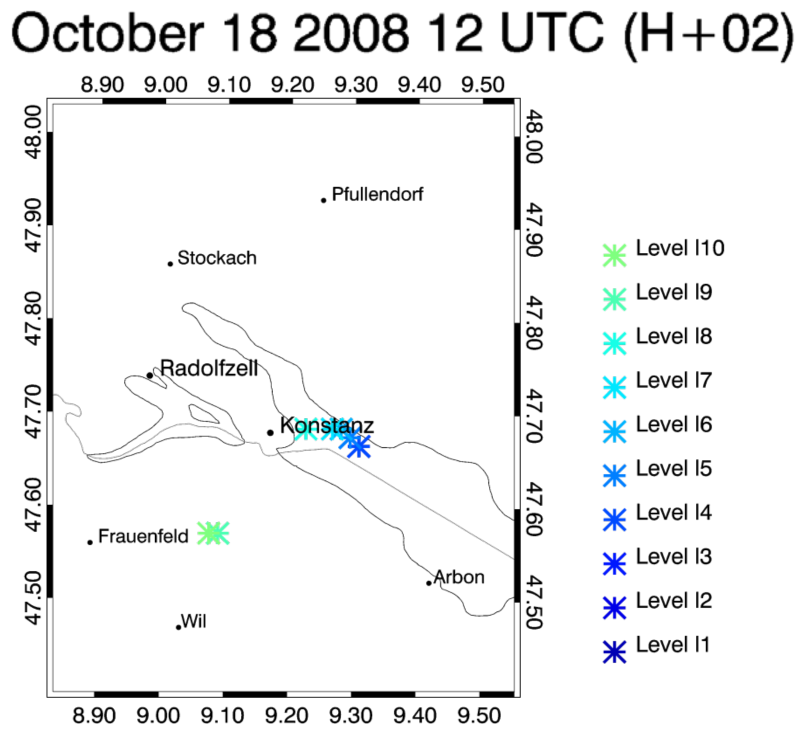
(from Nadine Goris, PhD thesis 2011)

What is important to observe for O_3 prediction?

δO_3 > δHCHO > δCO > δHONO > δOH

with NO and NO_2 typically varying between $\delta O_3 > \delta NO_x > \delta$HONO dependent on the specific chemical scenario.





Orders of magnitude of different compounds (left) and position of maximal values (right) of maximal singular vector entries per model level.

Where is it important to observe for O_3 prediction?

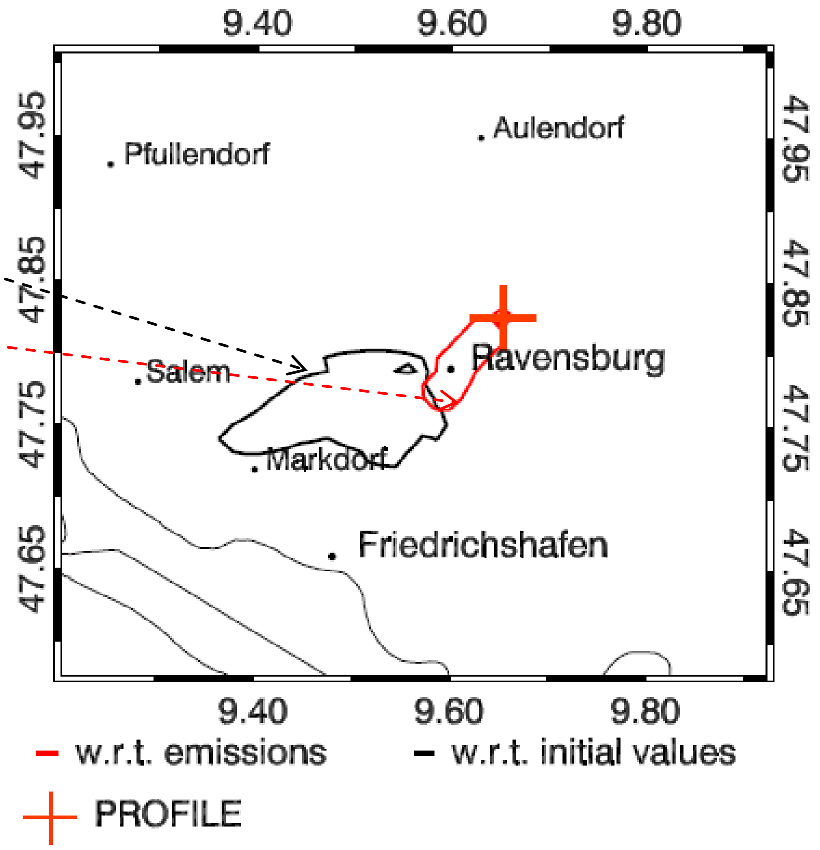
Example of maximal sensitivity contours of SV for

- HCHO initial values (black) and
- emission rates (red) for flight

19.Oct. 2008, t_I=14:00, t_F=15:20 at

+ for final location.

Emission sensitivity is concentrated in the Ravensburg urban area.



RESULTS

Implementation of emission rate optimisation

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

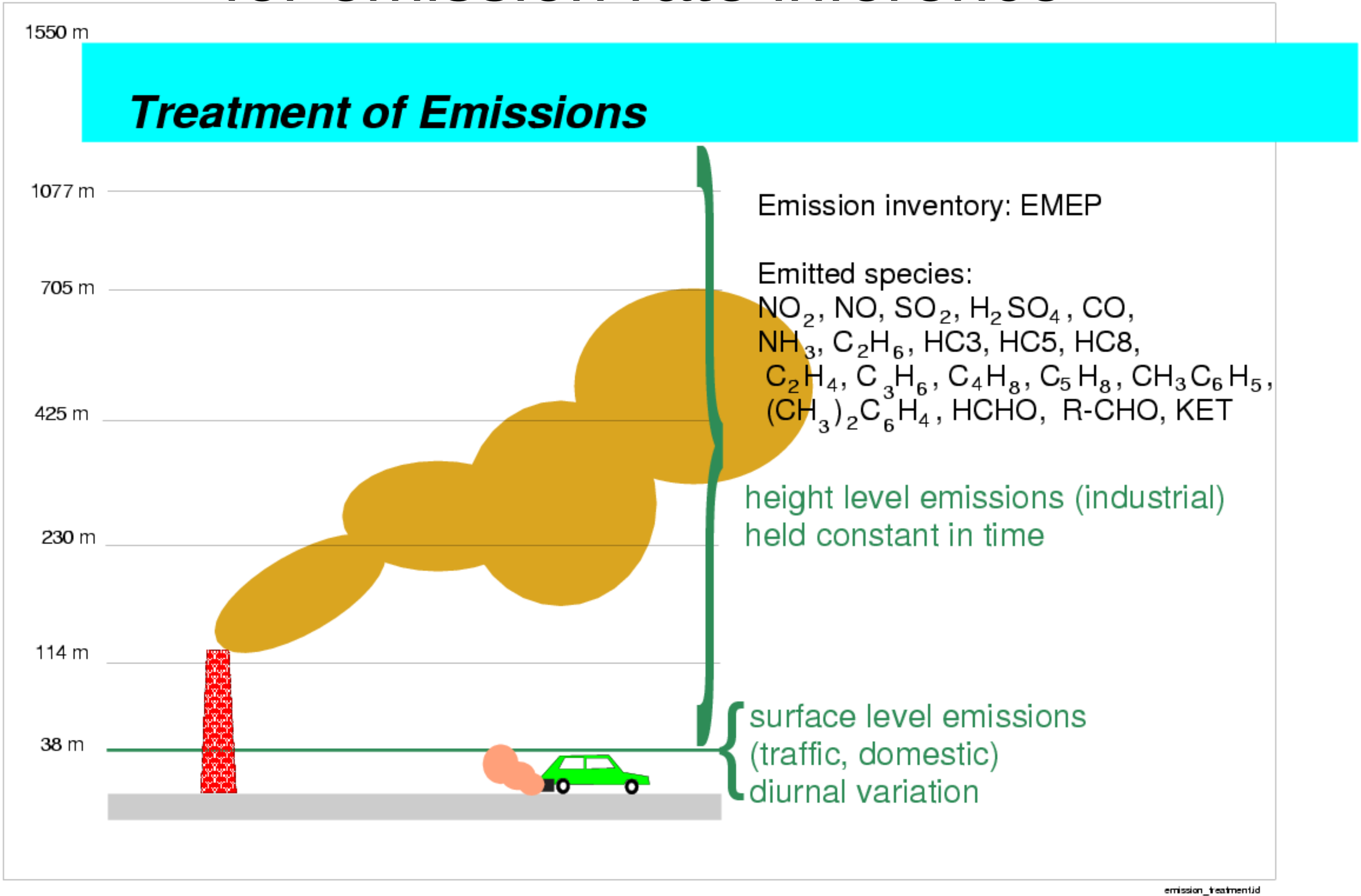
Emission Rate Optimization

minimize cost function

$$
\begin{aligned}
J(\mathbf{x}(t_0), \mathbf{e}) =& \frac{1}{2}(\mathbf{x}^b(t_0) - \mathbf{x}(t_0))^T \mathbf{B}_0^{-1} (\mathbf{x}^b(t_0) - \mathbf{x}(t_0)) + && \text{deviations from background initial state} \\
& \frac{1}{2}\int_{t_0}^{t_N} (\mathbf{e}_b(t) - \mathbf{e}(t))^T \mathbf{K}^{-1} (\mathbf{e}_b(t) - \mathbf{e}(t)) dt + && \text{deviations from a priori emission rates} \\
& \frac{1}{2}\int_{t_0}^{t_N} \left(\mathbf{y}^0(t) - H[\mathbf{x}(t)]\right)^T \mathbf{R}^{-1} (\mathbf{y}^0(t) - H[\mathbf{x}(t)]) dt && \text{model deviations from observations}
\end{aligned}
$$

$\mathbf{x}^b(t_0)$	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

Treatment of the inverse problem for emission rate inference



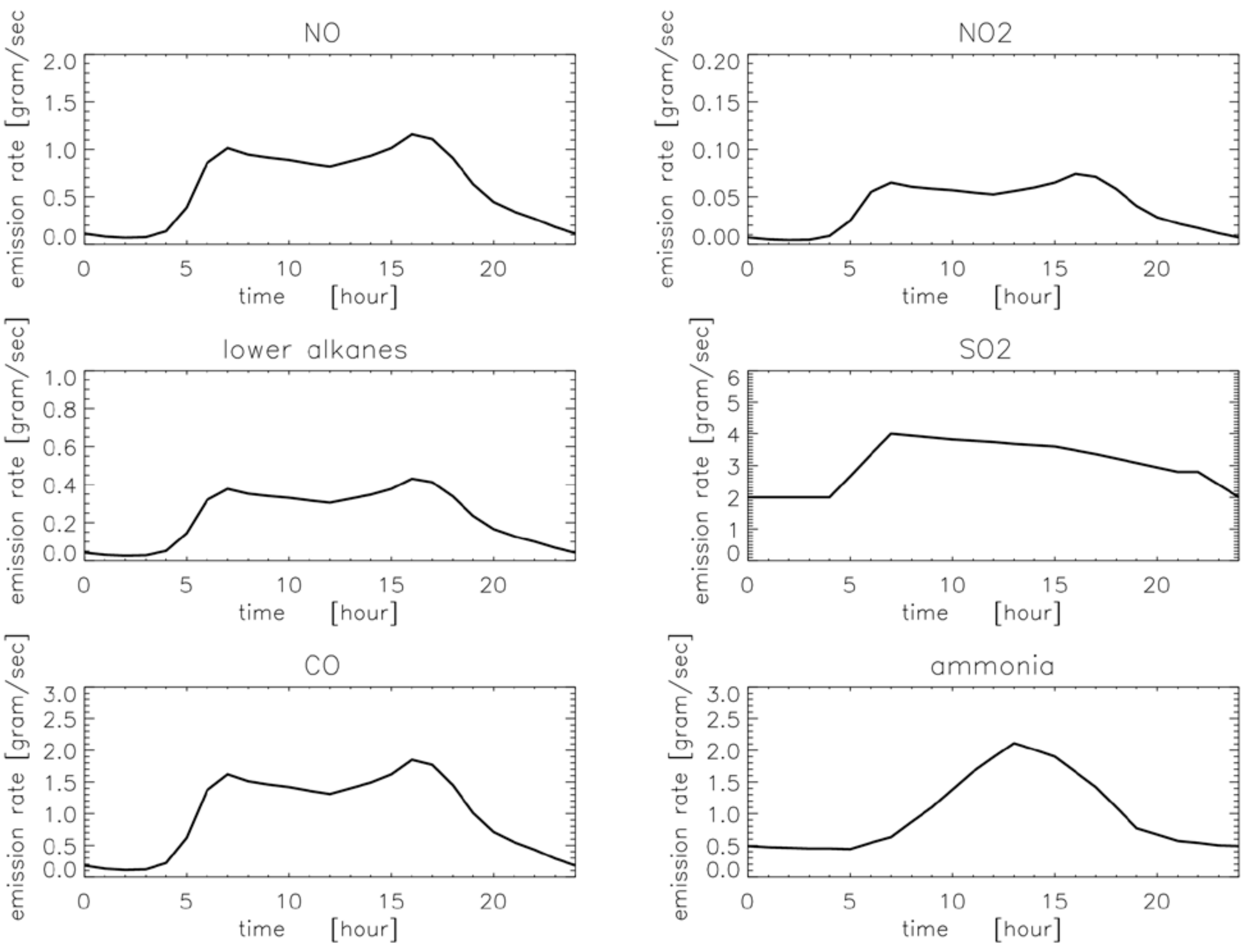
How can the degree of freedom of emission variation be reduced?

a "**strong constraint regularisation**"

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

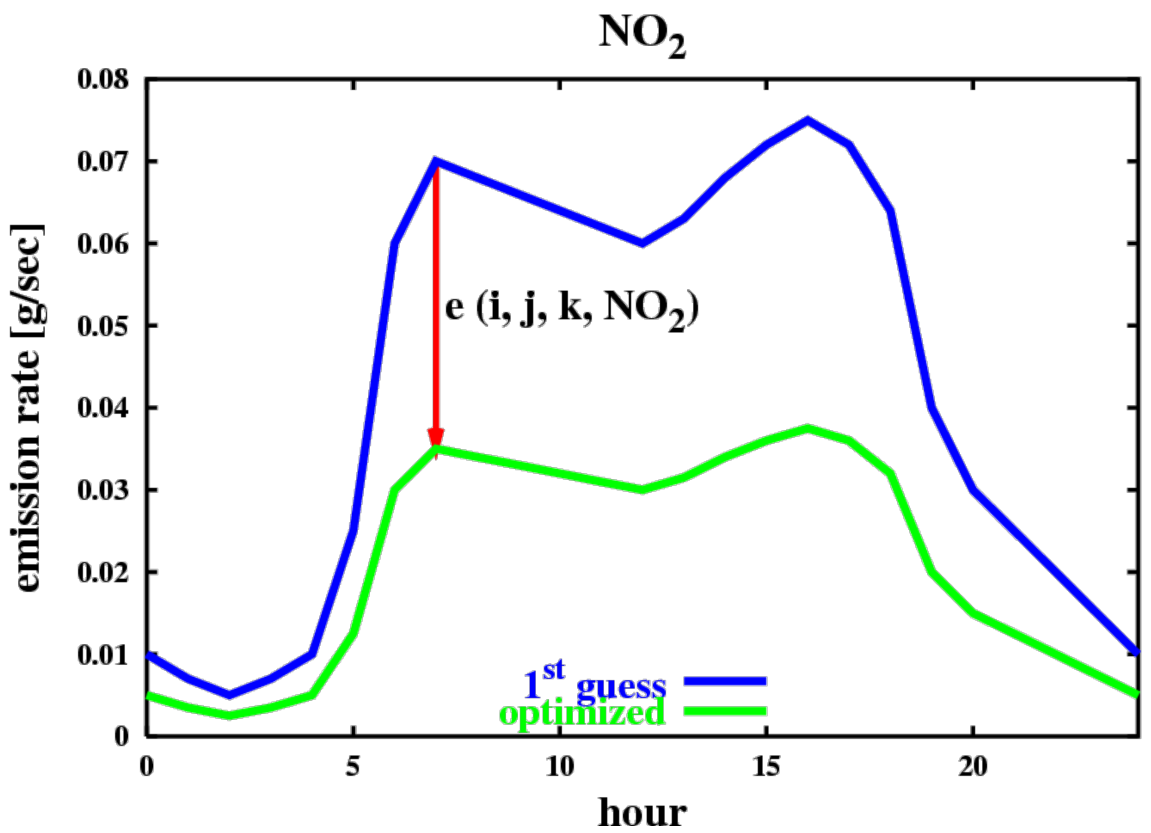
emission(t)=f(t;location,species,day) * v(location,species)

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



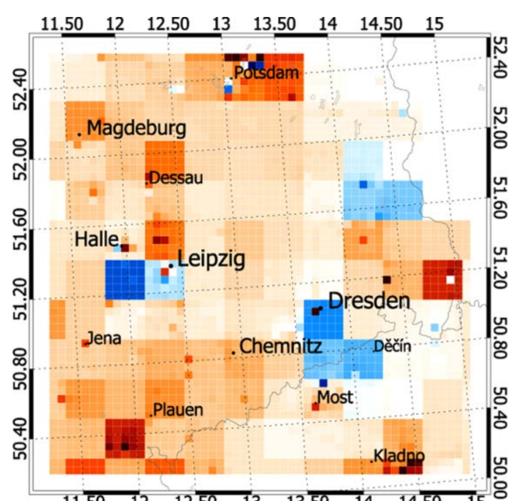
Optimisation of emission rates

amplitude optimisation for each emitted species and grid cell



Novel setup of joint emission rate-initial value optimisation
after upgrade of regional emission inventory

- Preparation:
  - adaptation to extended emitted species, („adjoint emission sources“)
  - more biogenic emission



	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 %

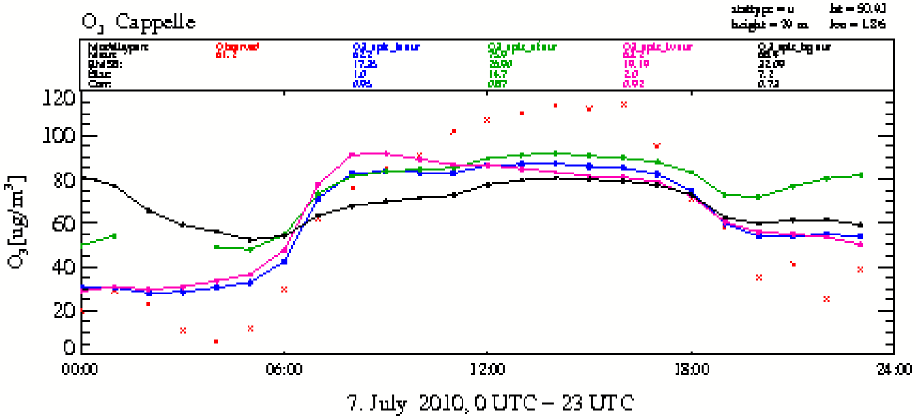
4D-var optimisation EURAD-IM

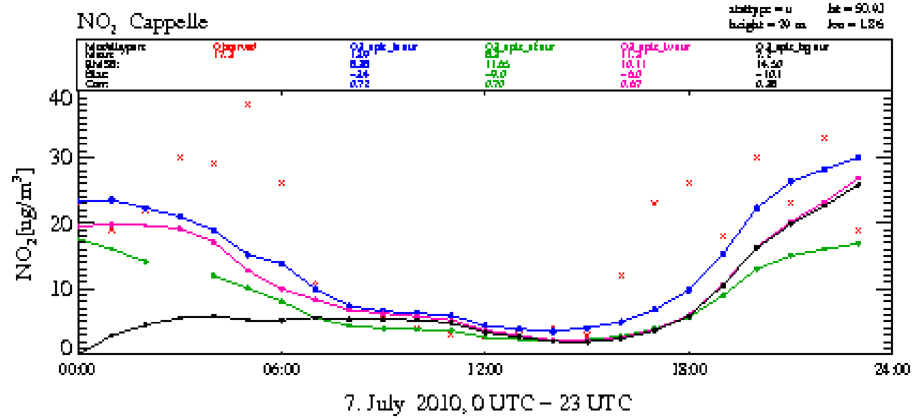
24 h window

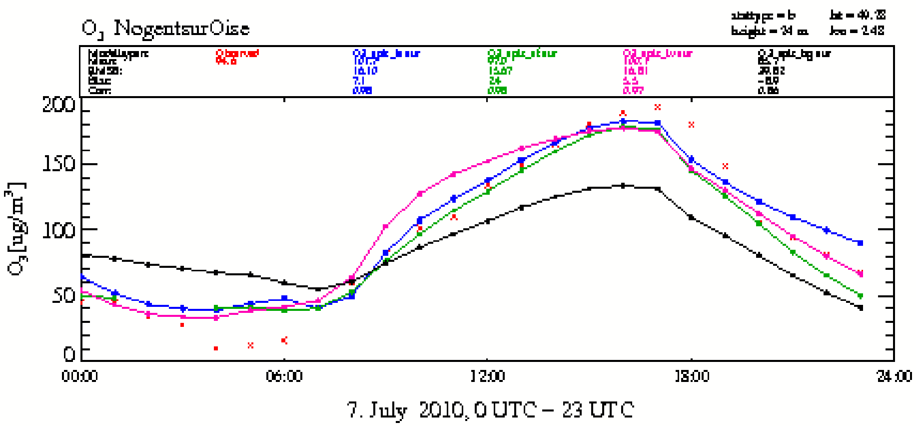
background (no assim)
initial values only
emission rates only
joint emi-ini. values

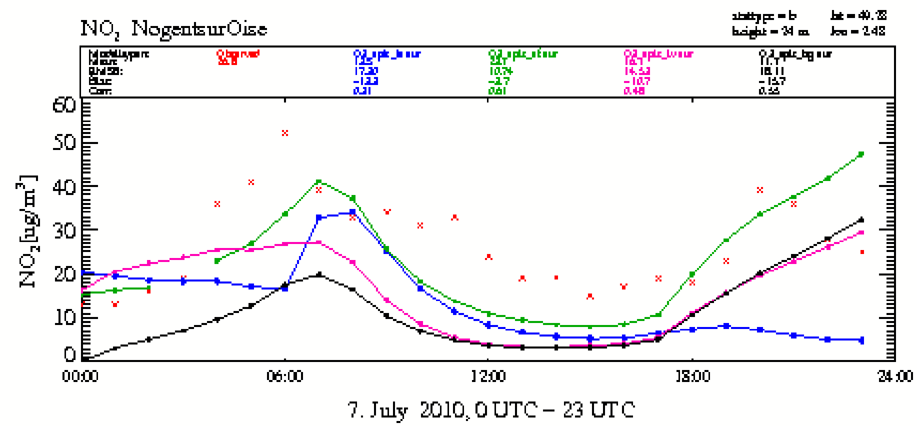
A potential constraint: consider O_x-concept [Guicherit 1988]:

$$O_x = NO_2 + Ozon = const$$



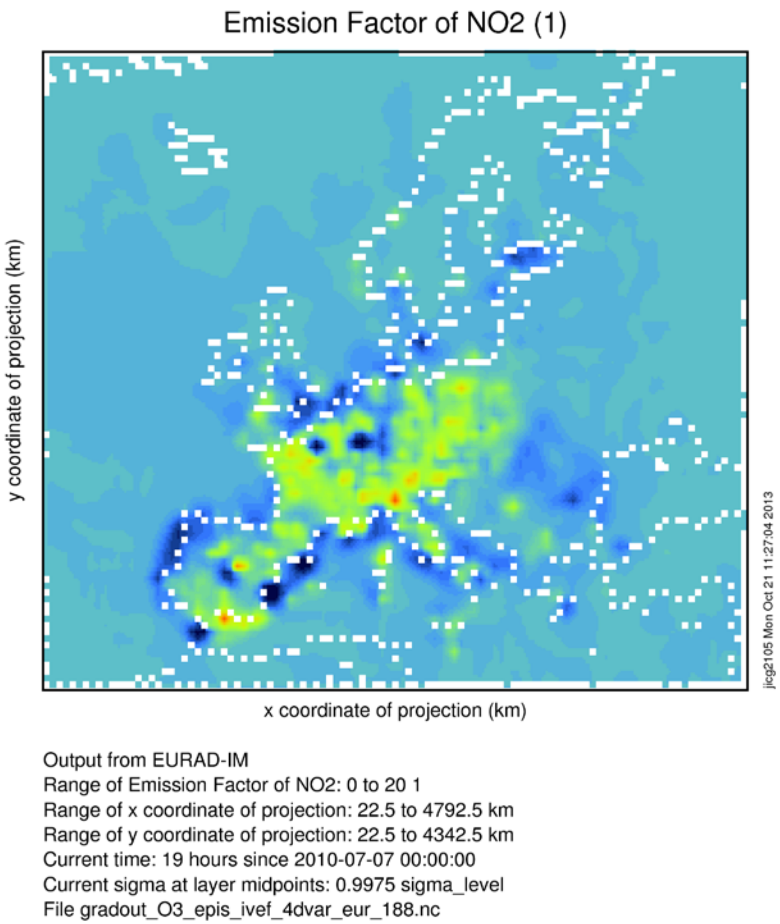




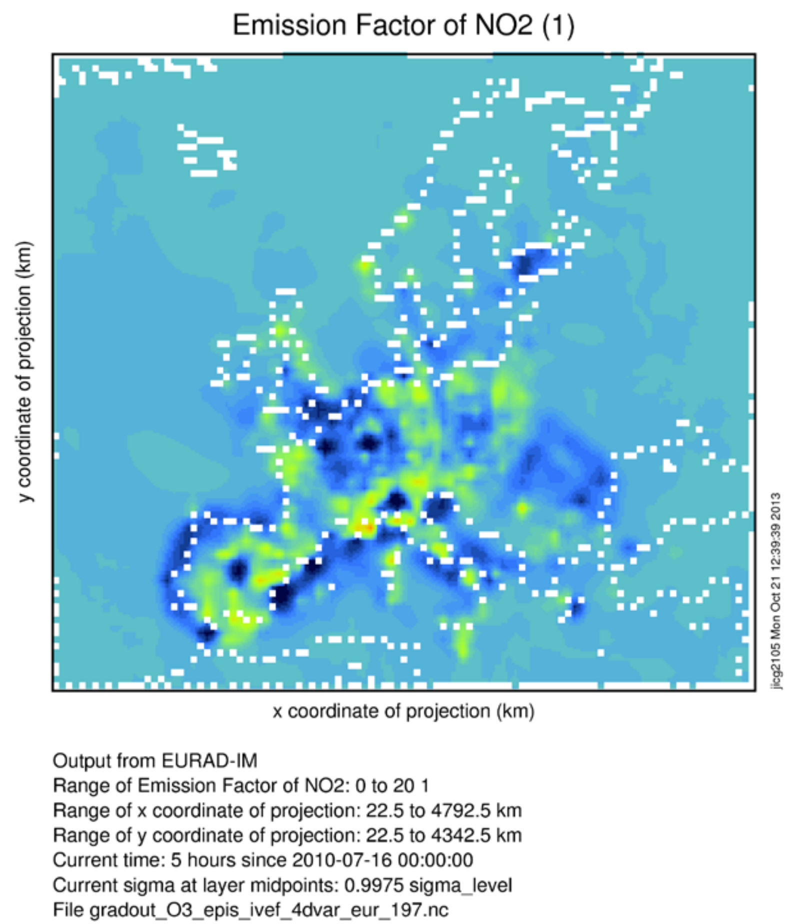


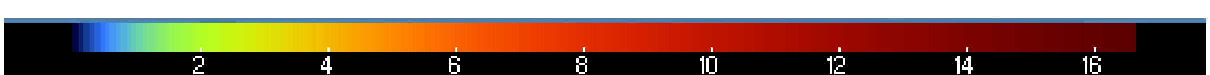
NO_2 emission correction factors summer 2010

7. July



16. July

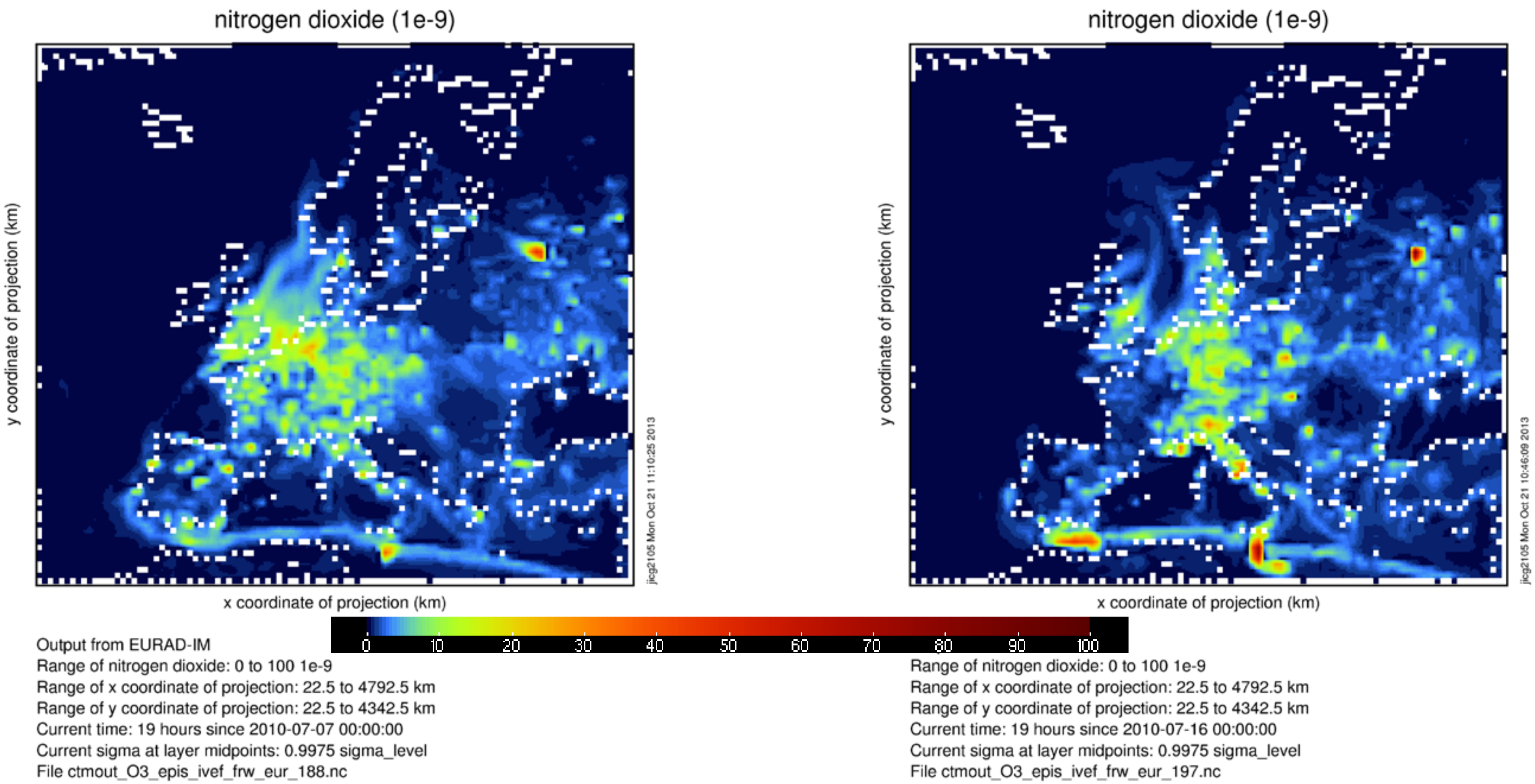




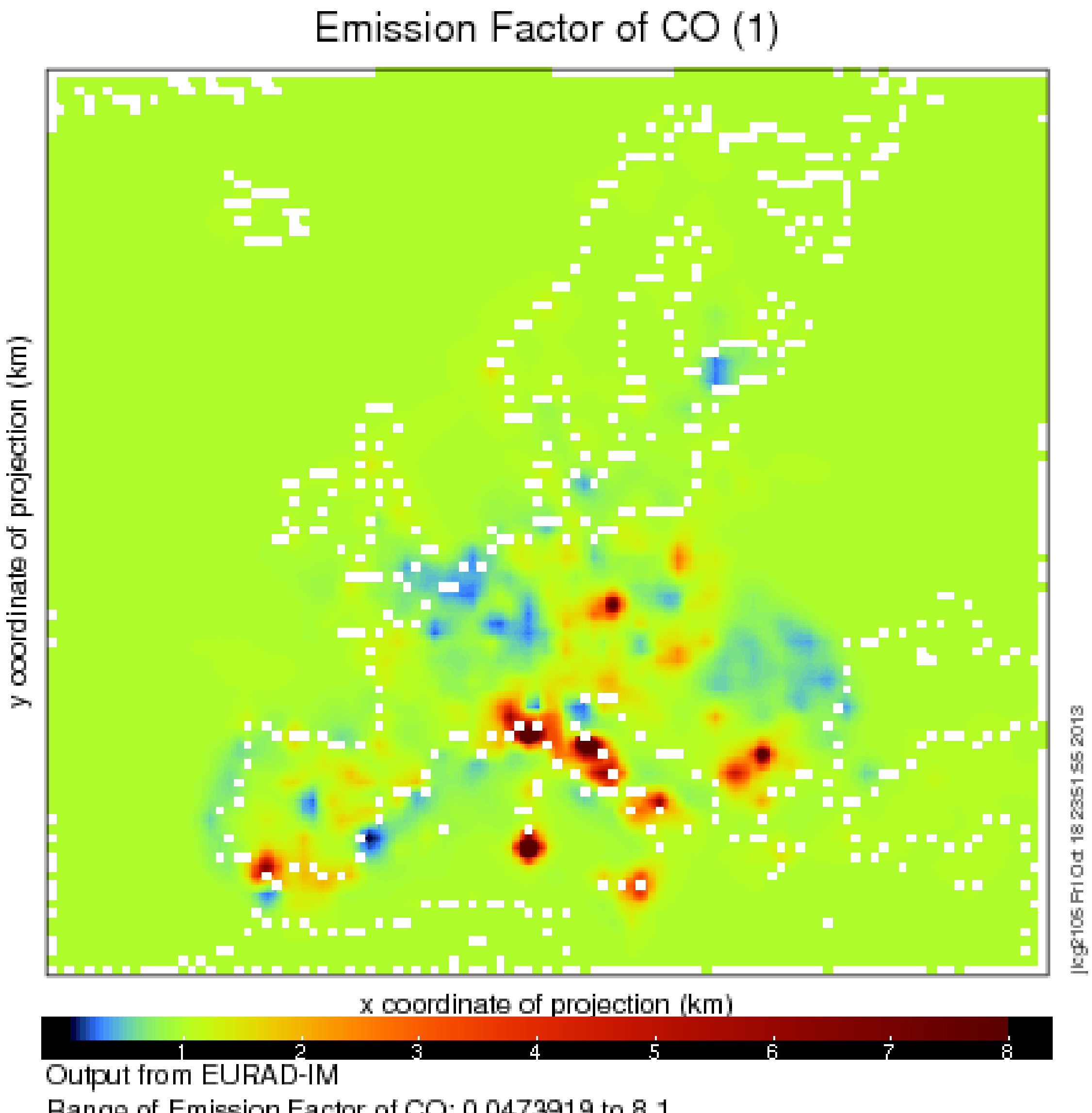
analyses ofNO_2 concentrations summer 2010

7. July

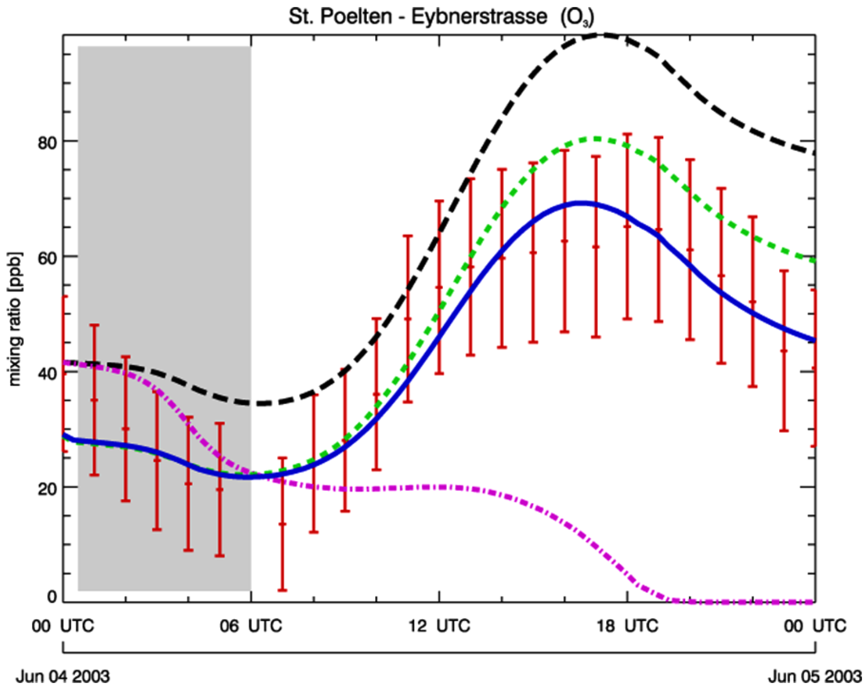
16. July



CO emission correction factors

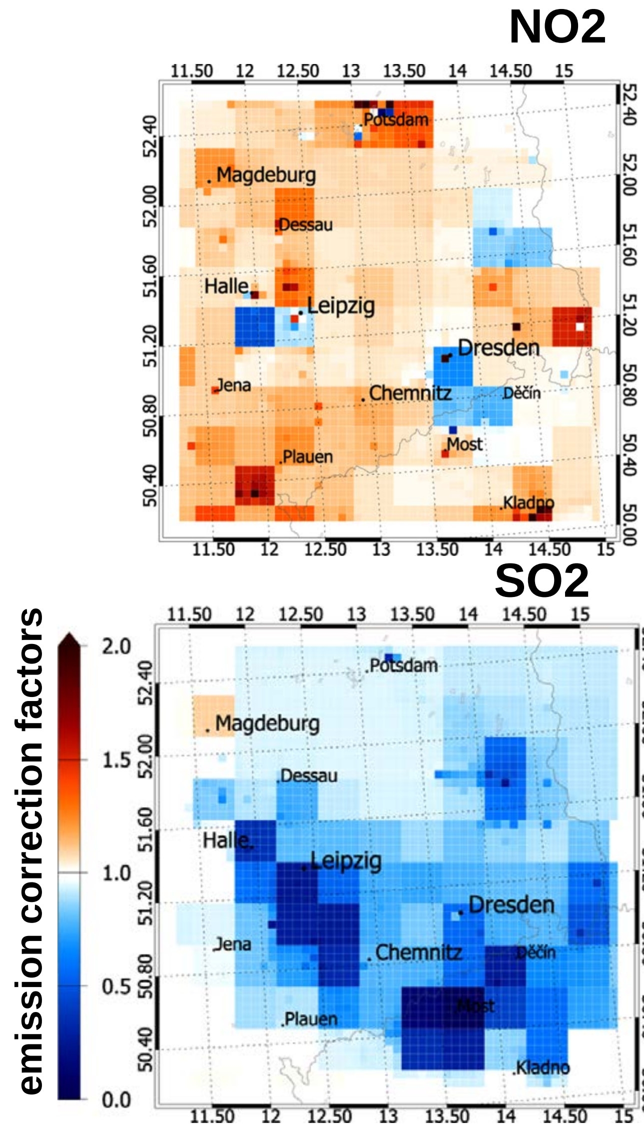


Analyses, Example (i):
Analysis of emissions by 4D-var (VERTIKO)



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

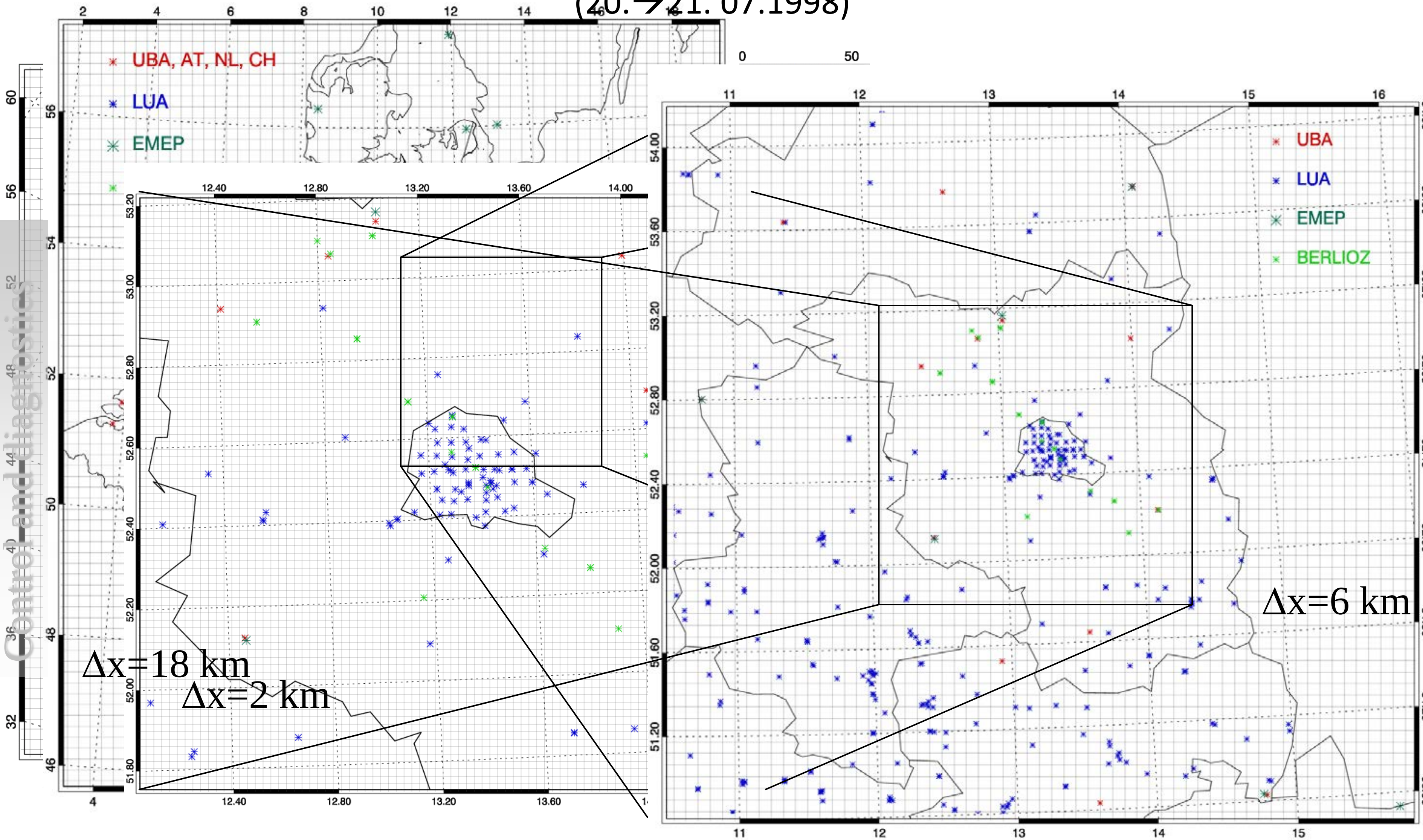
- - - - -	Control run without data assimilation.
······	initial value optimisation.
-·-·-·-·-·	emission factor optimisation.
———	joint initial value and emission factor optimisation (Strunk et al., 2011)



Which is the requested resolution?

BERLIOZ grid designs and observational sites

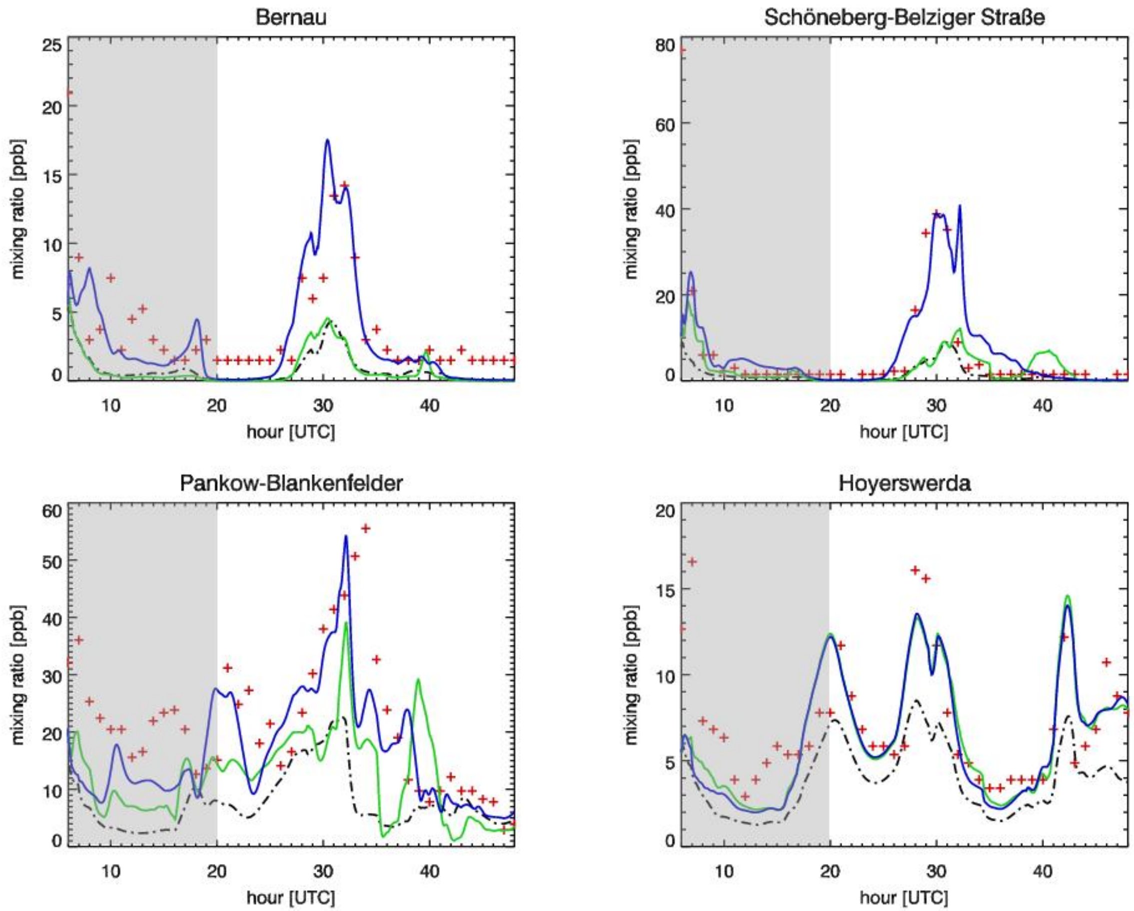
(20.→21. 07.1998)



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

NO

NO_2



Time series for selected NOx stations on nest 2.

+ observations,

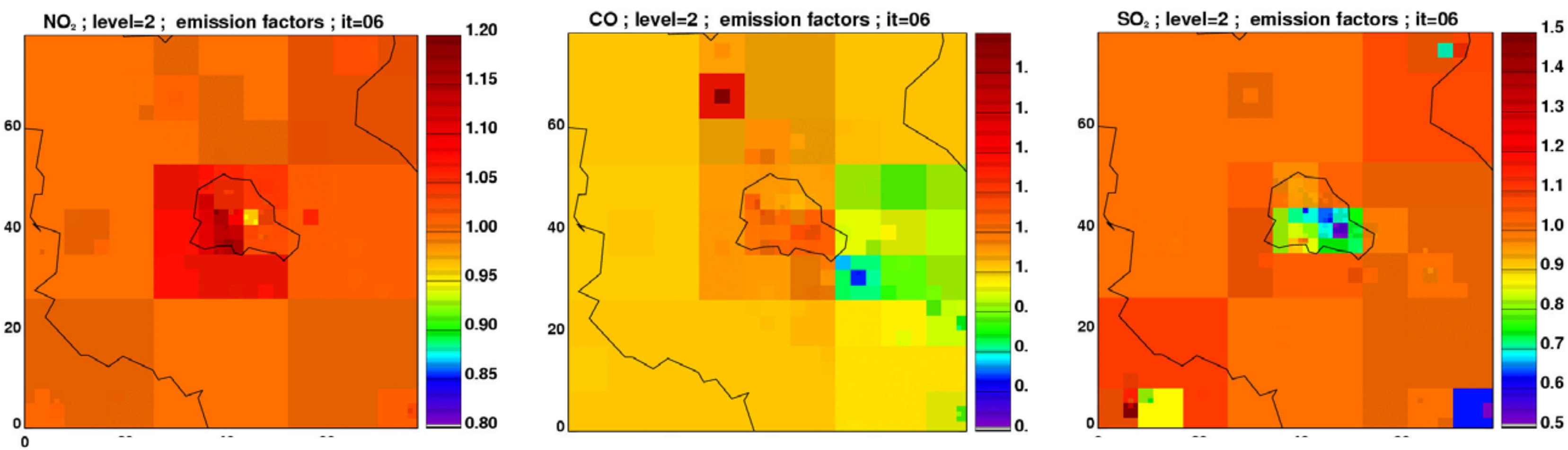
-- - no assimilation,

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

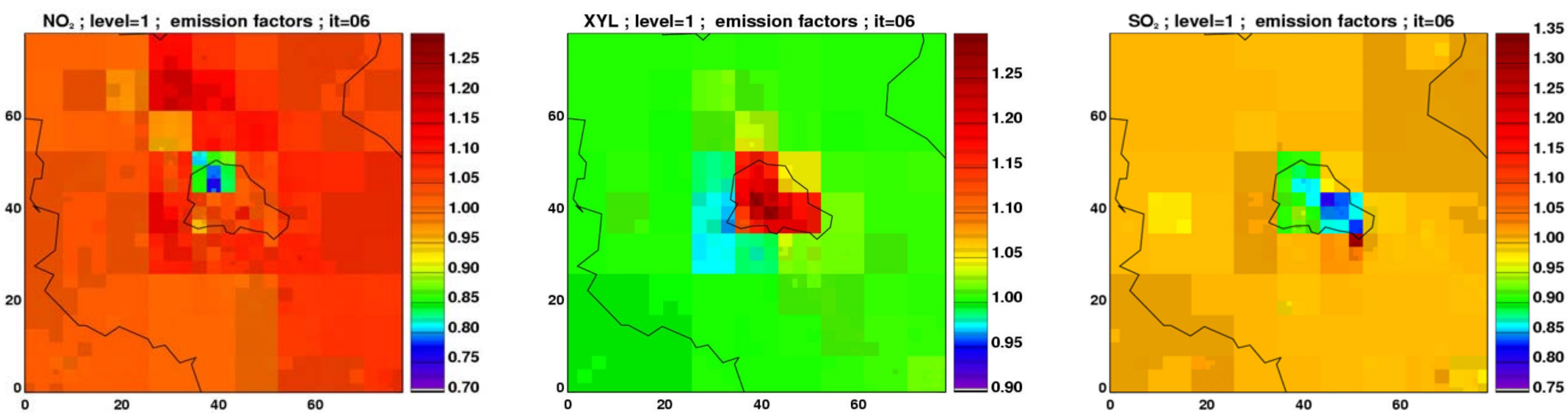
-_____N2 assimilation.(**6 km**),

-grey shading: assimilated observations, others forecasted.

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



NO_2, (xylene (bottom), CO (top)) SO_2 .



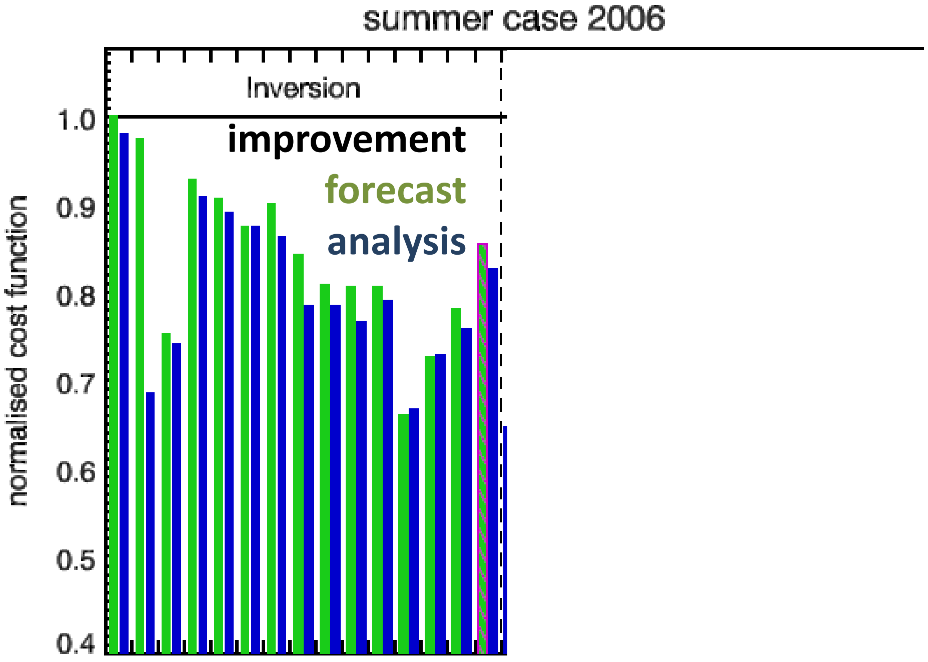
height layer ~32-~70m

surface

OMI NO2 column assimilation: Forecast validation summer (high convection)/ winter (low inversion)

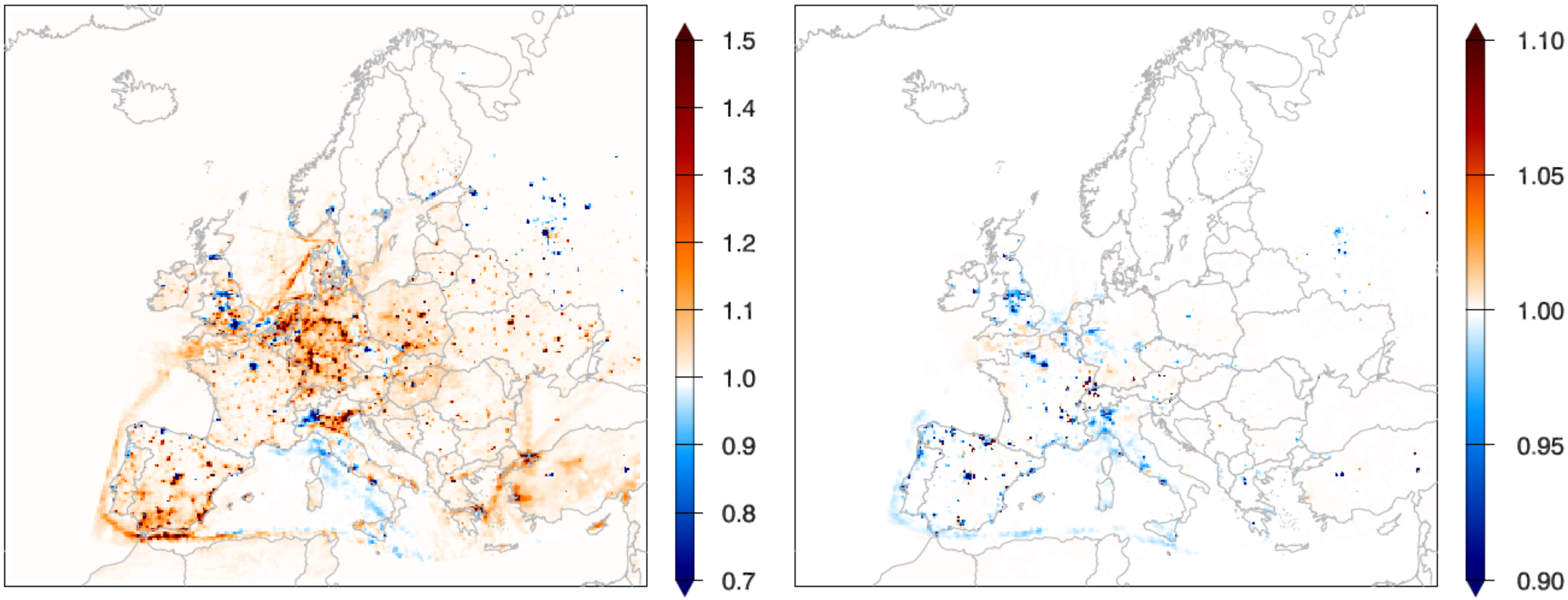
July 1st to 15th 2006 inversion
16th-31st validation (by SCIA)

December 11 to 25, 2007 inversion
26th to Jan. 10th 2008 validation



Cost function values of the background simulation (green) and analysis results (blue), normalised by the cost function value of the control simulation without data assimilation

NO2 Emission factors TNO emission inventory

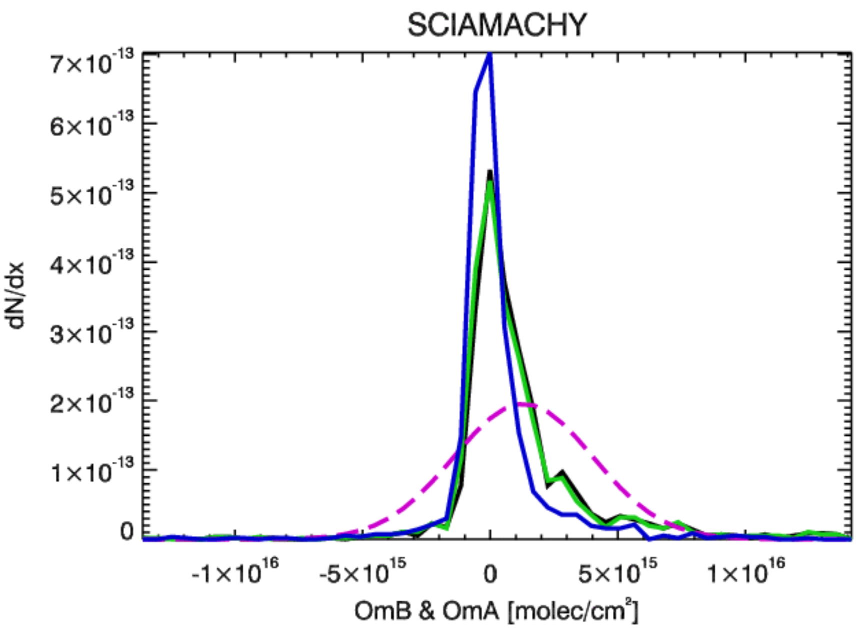


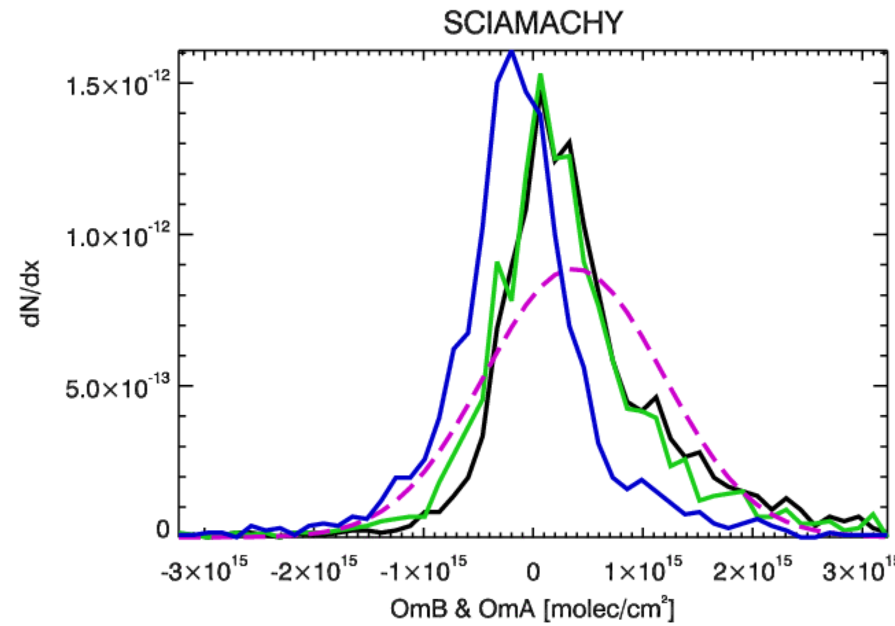
Comparison of nitrogen dioxide emission factor results for the summer case study (left panel) and the winter episode (right panel). Please note the different ranges on the color scale.

SCIAMACHY

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

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





Control run (OmC) (no data assimilation at all,) black bold line,

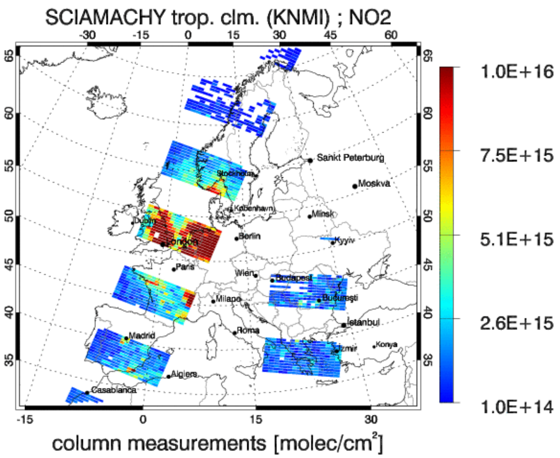
forecasted values (OmF) green bold line,

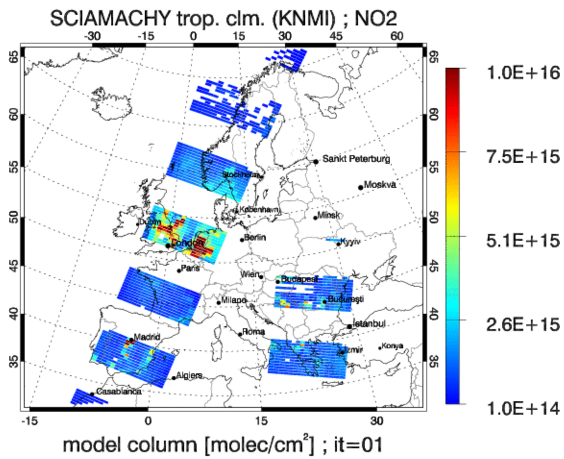
analyses (OmA) blue bold line.

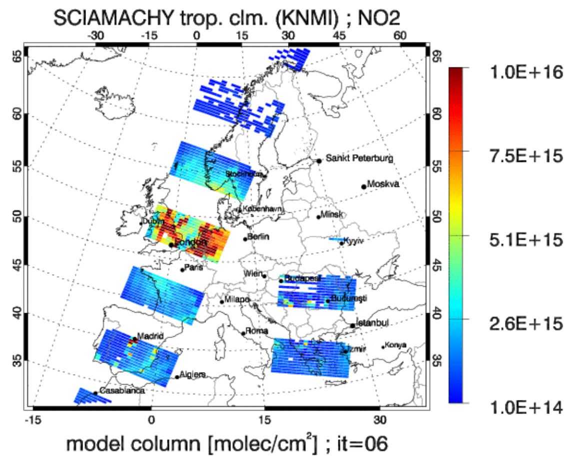
For comparison: Gaussian fit to OmF pdf by

mean and standard deviation given by broken purple line.

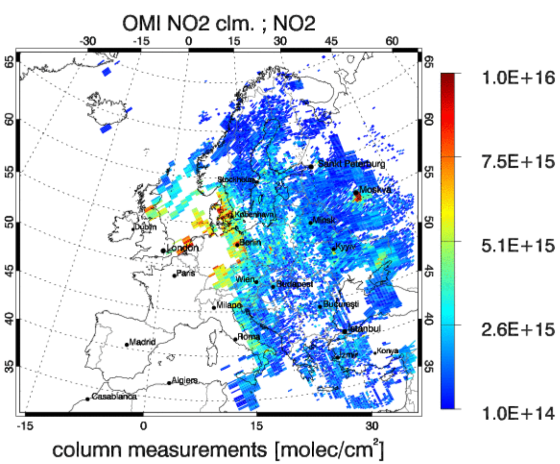
Comparison of NO2 tropospheric columns in molecules/cm2 for July 6th, 2006, 09-12 UTC.

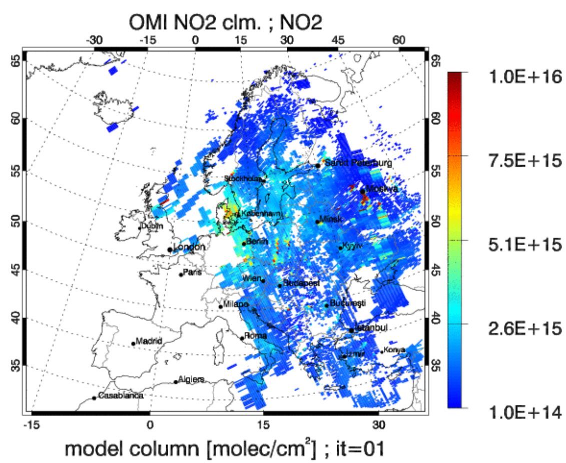


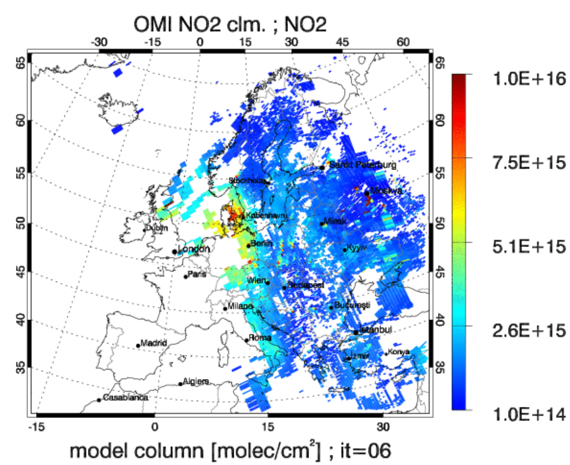




assimilated values ($\mathbf{y}$); :EURAD forecasted ($\mathbf{Hx}_b$); column analyses ($\mathbf{Hx}_a$).

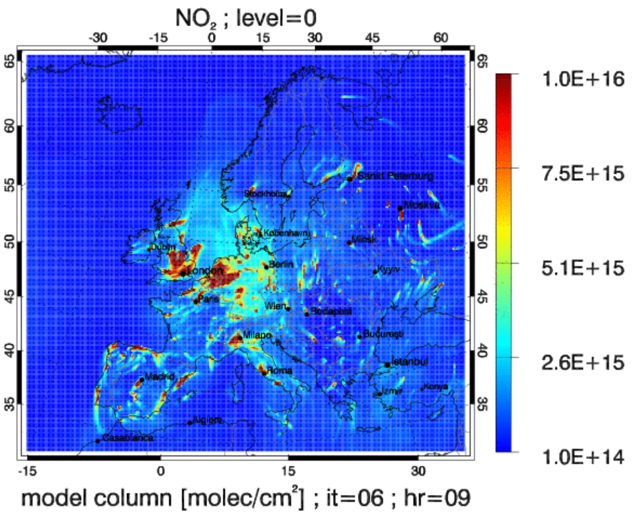




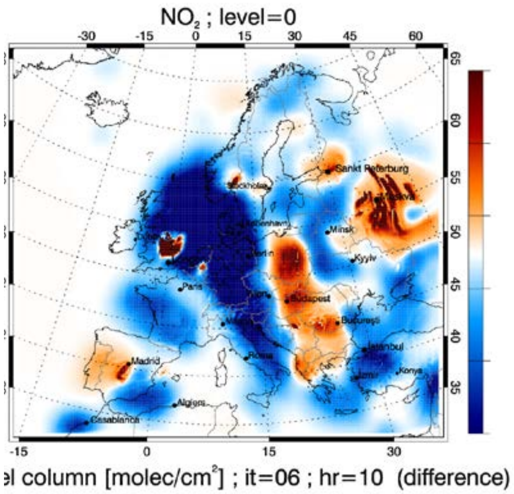


Data assimilation result in terms of tropospheric columns for July 6th, 2006. NO2 model columns based on OMI and SCIAMACHY

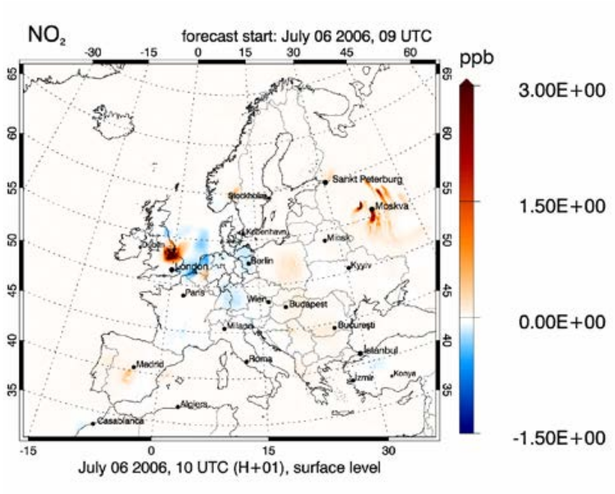
assimilation within interval, 09-12 UTC.



Analysed column densities

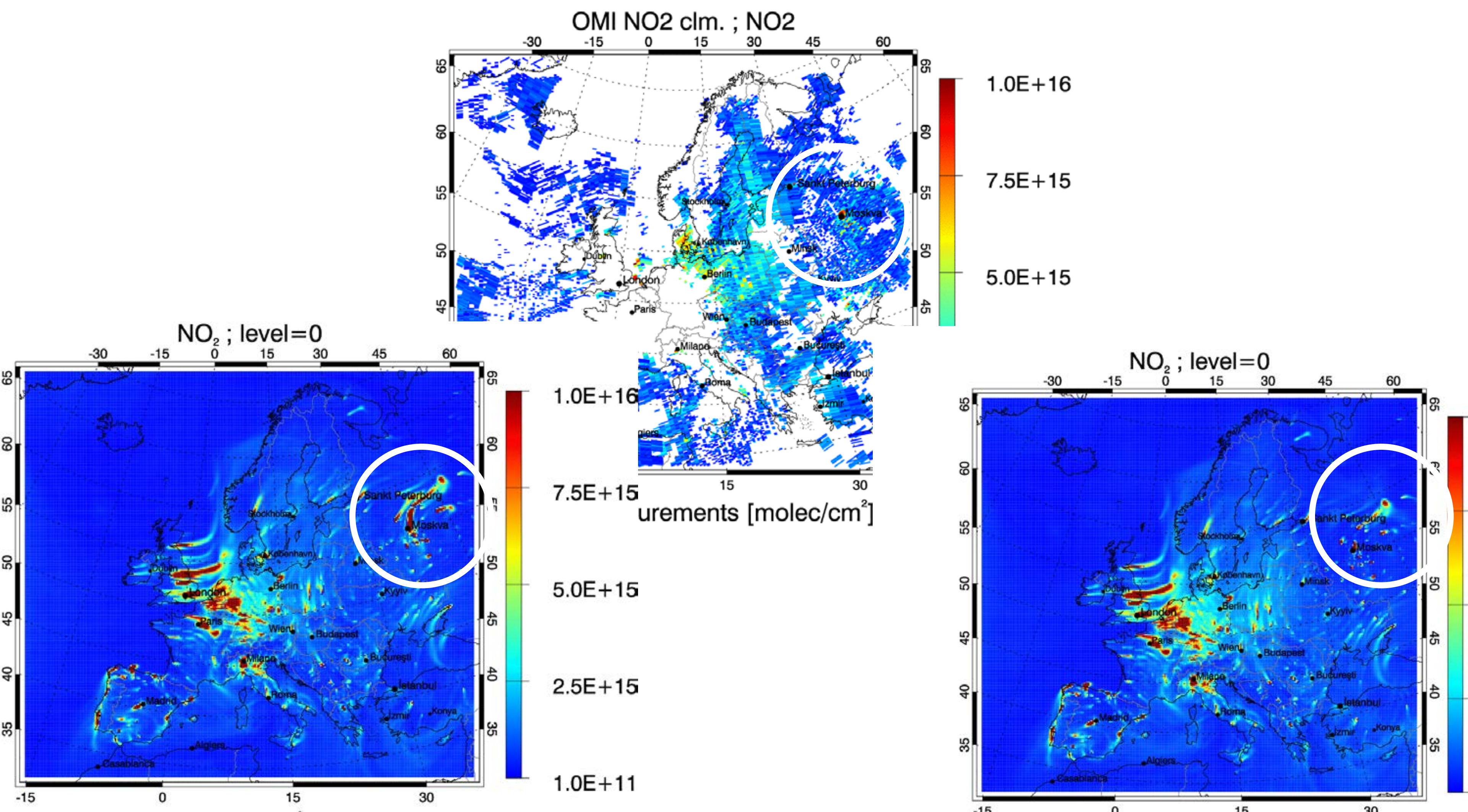


difference field from background



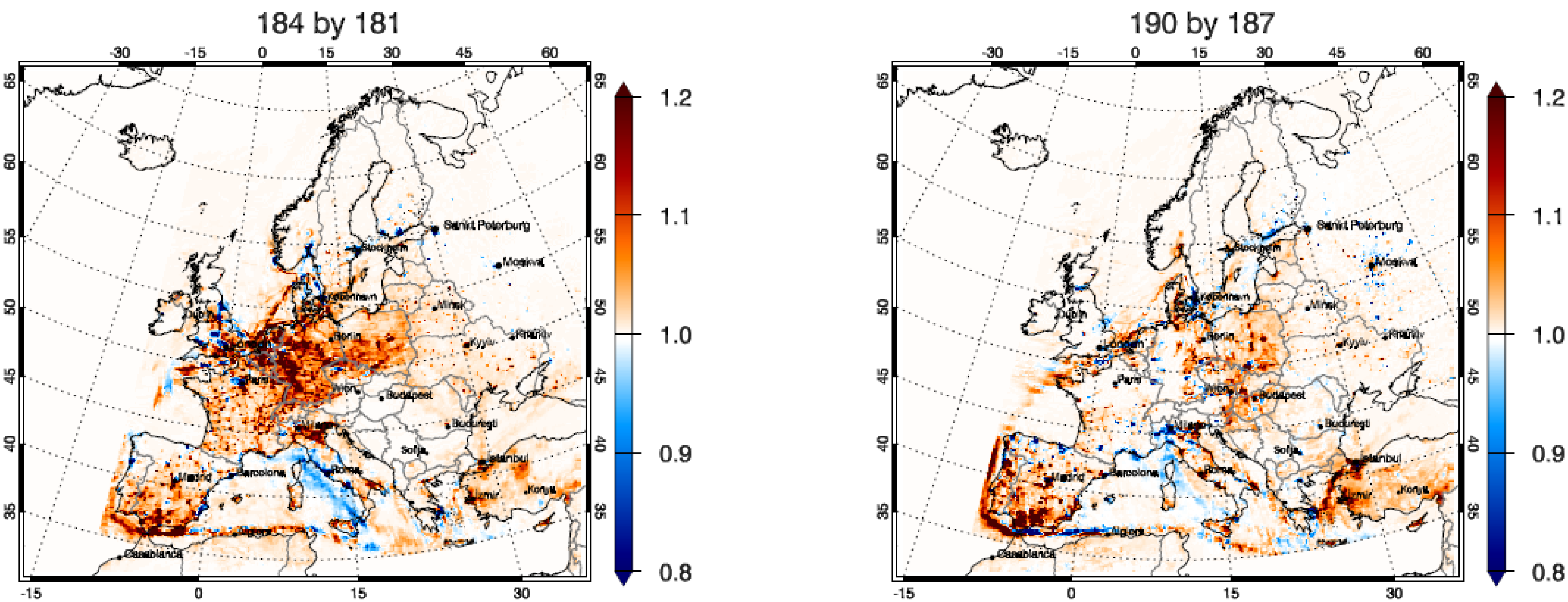
induced surface concentration changes by NO2 ppb

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

Summary

- Emission rate estimation cannot be performed isolated. Rather, other controling parameters (initial values) must be included.
- A smoother type spatio-temporal data assimilation algorithm is required (integration „backward in time and process“: 4D-var)
- As ill-conditioned problem, regularisation is needed.
- Future: chemial scenario dependent covariances need to be designed
- A rigorous (control) theory based balance between initial value and emission rate weights is needed.
- Emission rates need be optimized SNAP-wise.

Some references

Elbern, H. A. Strunk, L. Nieradzik, Inverse modelling and combined state-source estimation for chemical weather, In: "Data Assimilation: Making sense of observations", Springer, eds. Lahoz, Khattatov, Menard, pp 491{513, doi 10.1007/978-3-540-74703-1 19, 2010.

Elbern, H., A. Strunk, H. Schmidt, and O. Talagrand, Emission rate and chemical state estimation by 4-dimensional variational inversion, ACP, 3749-3769, 2007.

Elbern, H., H. Schmidt, O. Talagrand, A. Ebel, 4D{variational data assimilation with an adjoint air quality model for emission analysis, Environ. Mod. Softw., 15, 539-548, 2000.

Goris, N., and H. Elbern Singular vector decomposition for sensitivity analyses of tropospheric chemical scenarios Atmos. Chem. Phys., 13, 5063-5087, 2013.