From columns to emissions:

What can we learn from satellite observations?

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MAX-PLANCK-INSTITUT
FÜR CHEMIE

From columns to emissions…

- Satellite measurements: NO_2 tropospheric columns V
- V depend on **emissions**, **transport**, and **chemistry**: $V=f(E,w,\tau)$
- "Mainstream" application: Inverse modelling
  - Tune **emissions** until modelled and observed columns match
  - Assumes that **transport** and **chemistry** are known!

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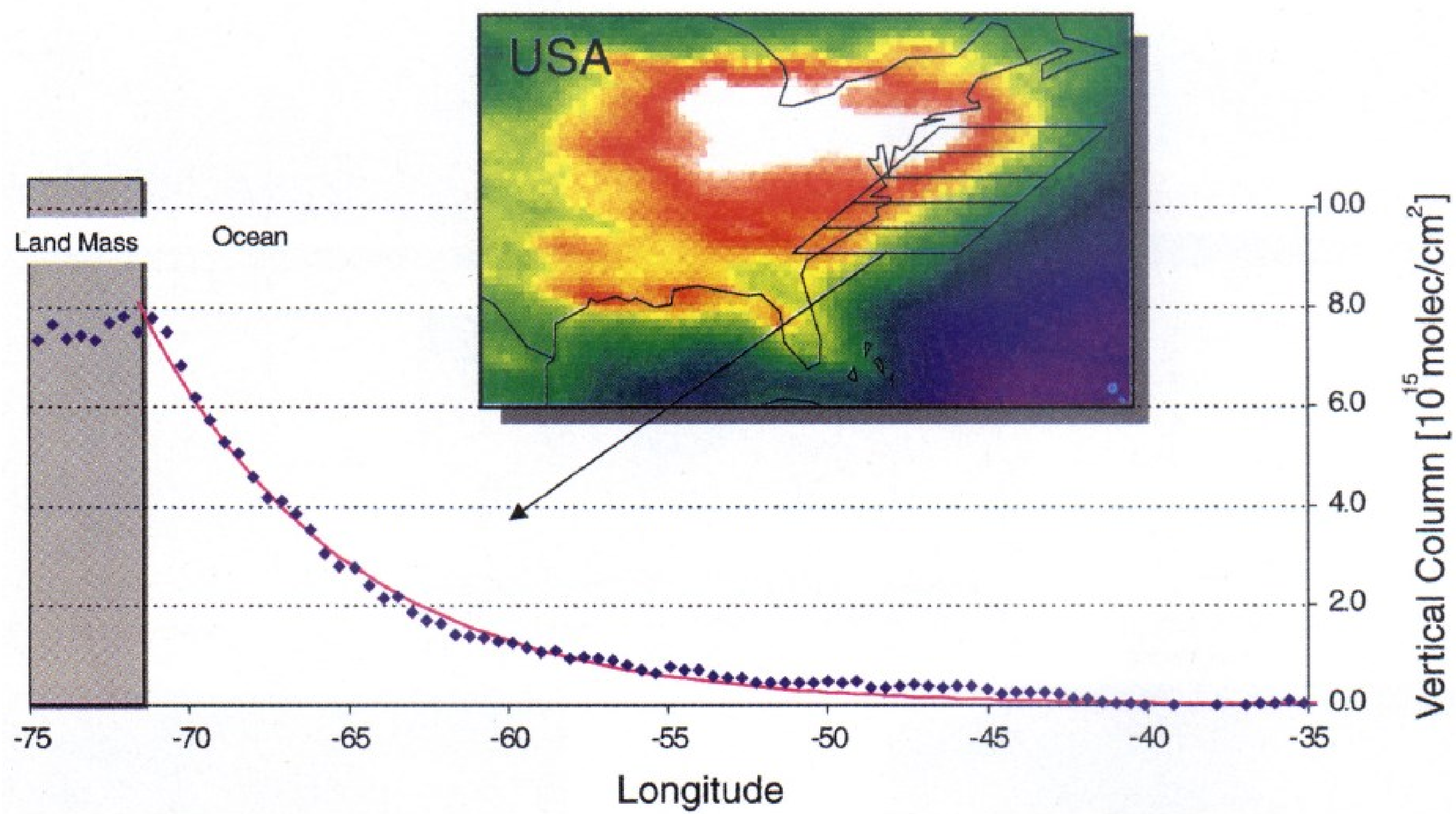
… and beyond:

- Downwind plume evolution contains **lifetime** information!
- Derive **E** independent from models

Estimates of NO_x lifetimes from satellite

- Leue et al., JGR, 2002:



- τ~1 d
- Biased high! (GOME-resolution!)

Estimates of NO_x lifetimes from satellite

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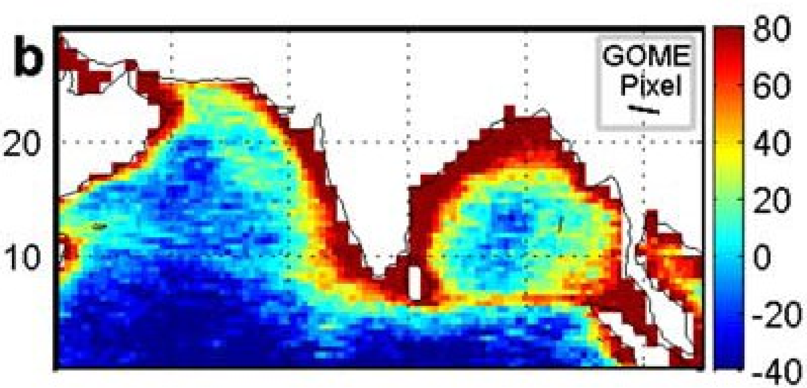
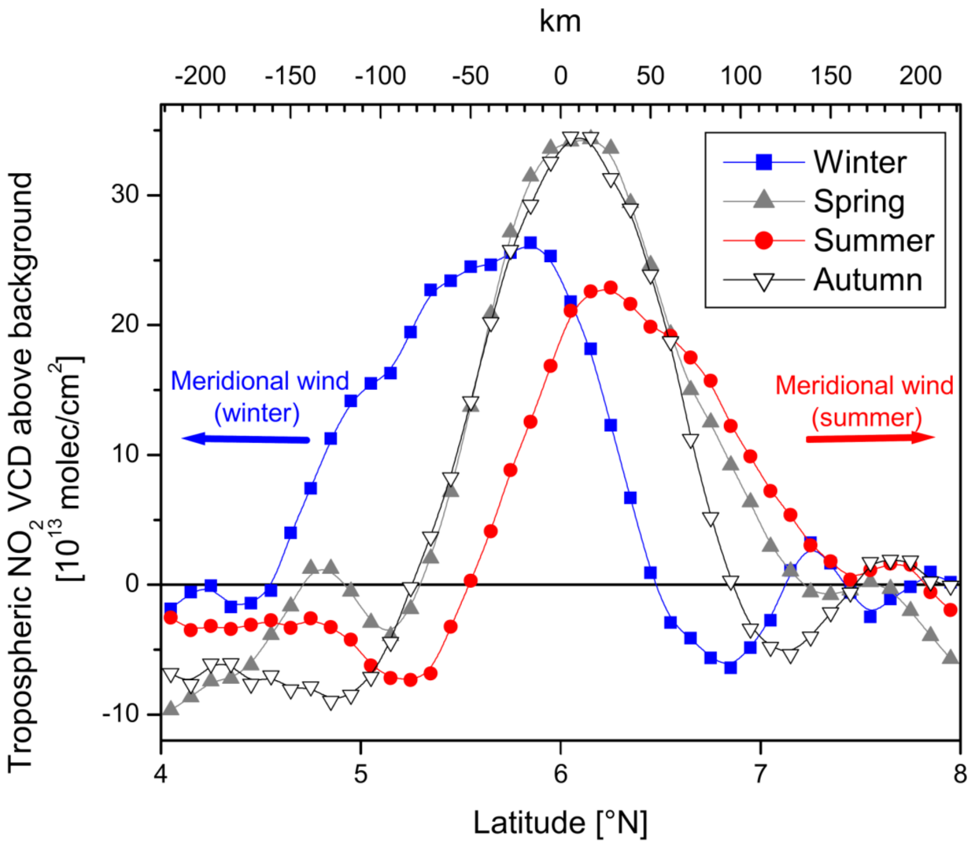
- Beirle et al., GRL, 2004:





- τ~5 h
- Specific conditions:
  strong & stable seasonal winds

Estimates of NO_x lifetimes from satellite

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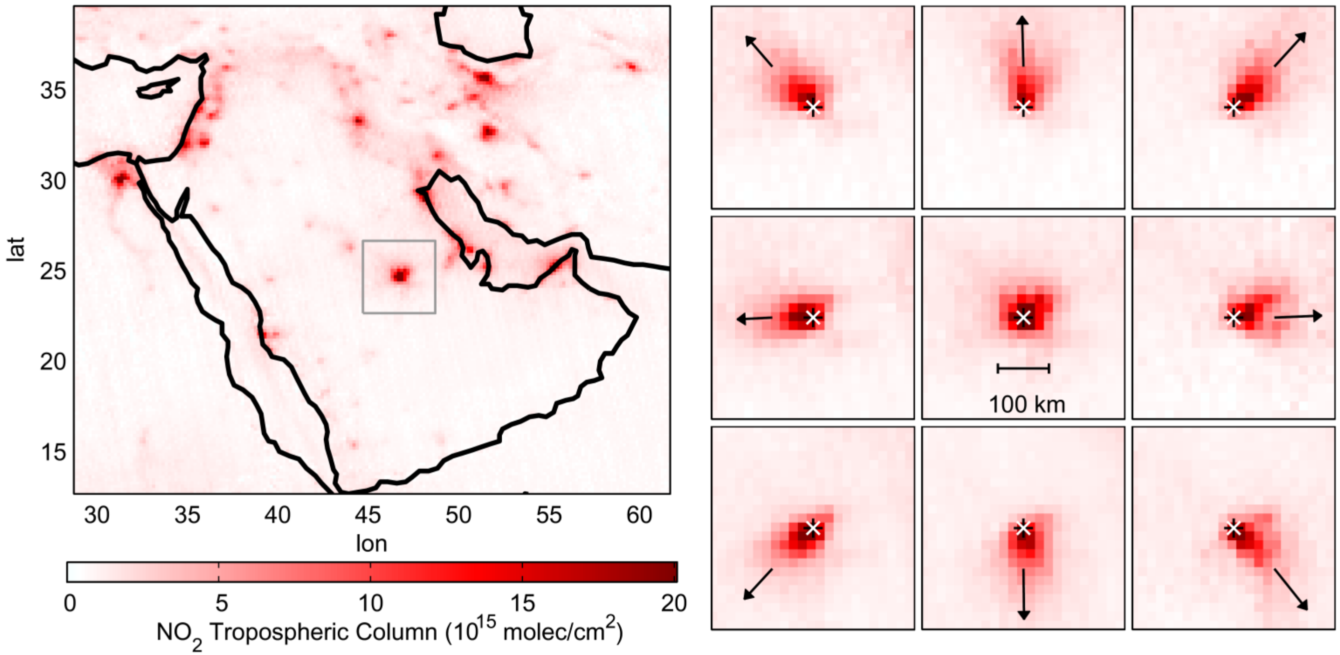
- Beirle et al., Science, 2011:



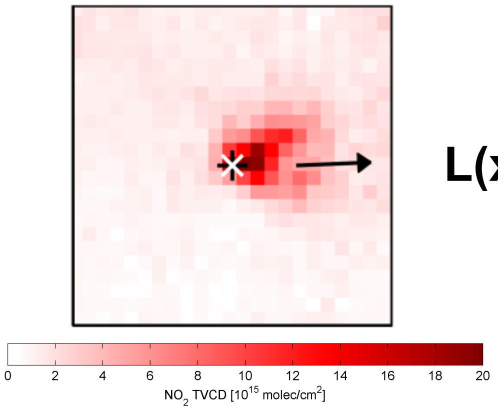
Mean tropospheric NO_2 TVCD 2005-2009 (DOMINO v1.02, cloud-free (TEMIS) For calm (<2m/s) and different wind directions (ECMWF)

Riyadh is particularly suited:

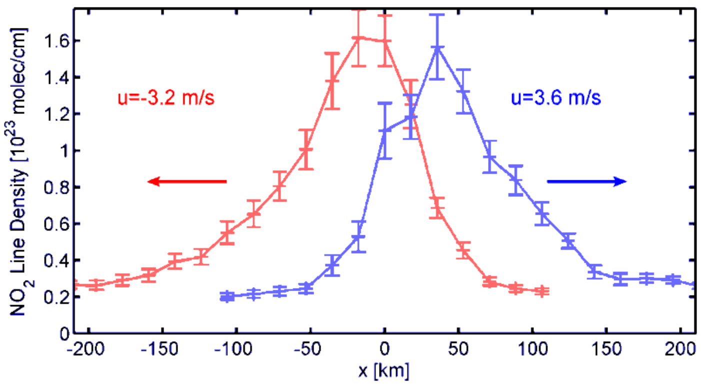
- **Strong, isolated** source
- **Homogeneous** terrain & wind fields (no coast!)
- **No clouds!**

Estimates of NO_x lifetimes from satellite

2D TVCD V(x,y) → 1D "Line density" (LD) L(x)



$L(x)=\int V dy$



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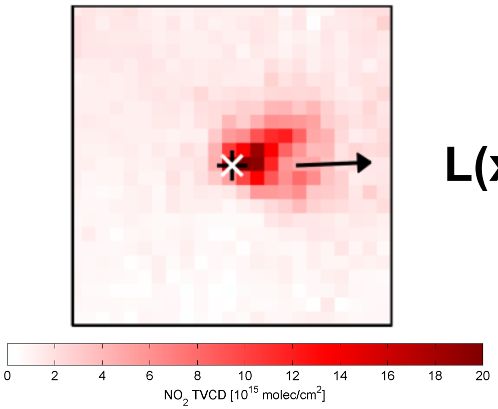
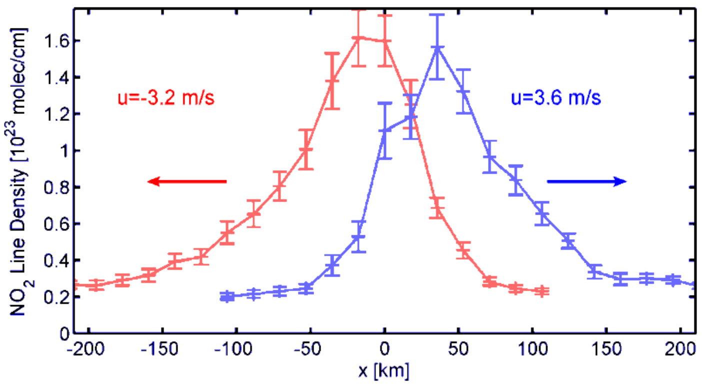
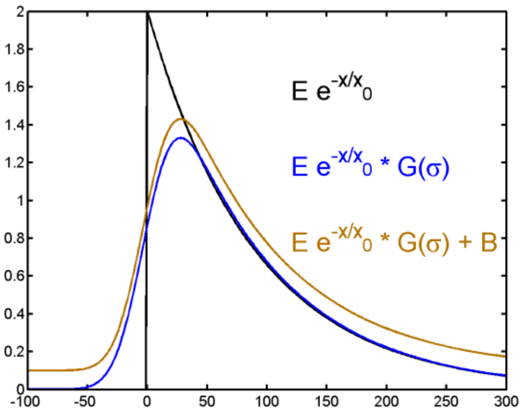


$L(x) = \int V dy$



Fit: Truncated exp



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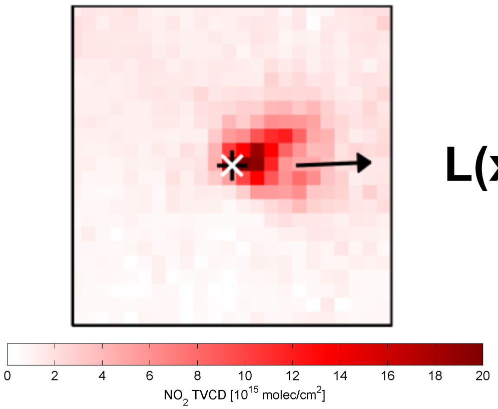

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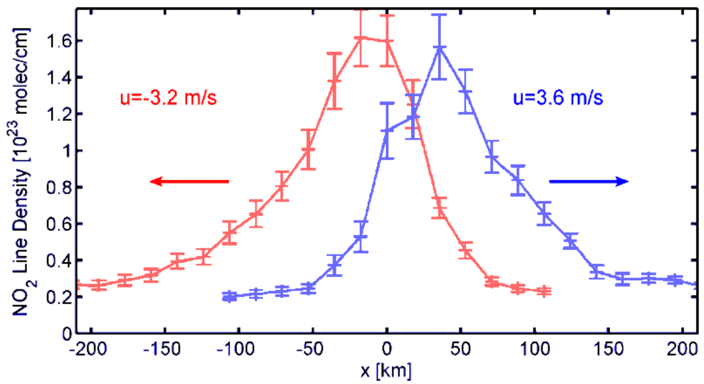
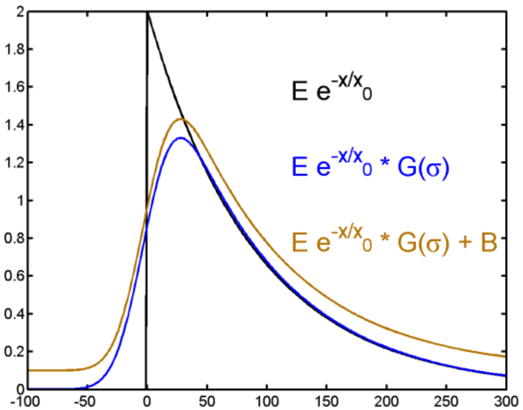


Fit: Truncated exp convolved with Gaussian



Gaussian accounts for
- spatial source extension
- dilution in wind direction
- changing wind speeds
- satellite ground pixel size

Estimates of NO_x lifetimes from satellite

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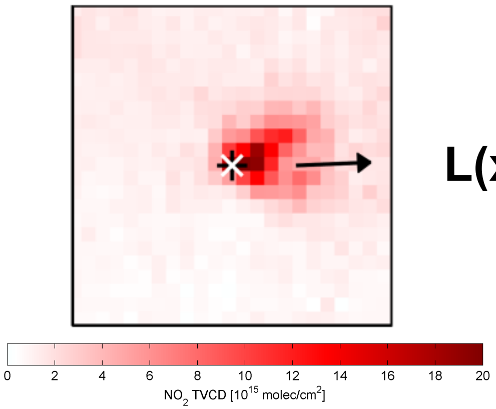

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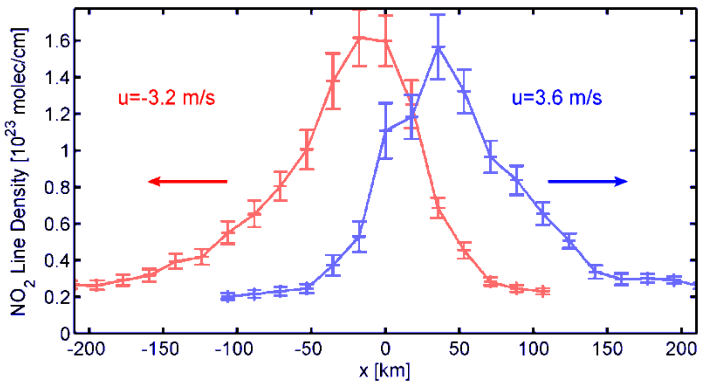
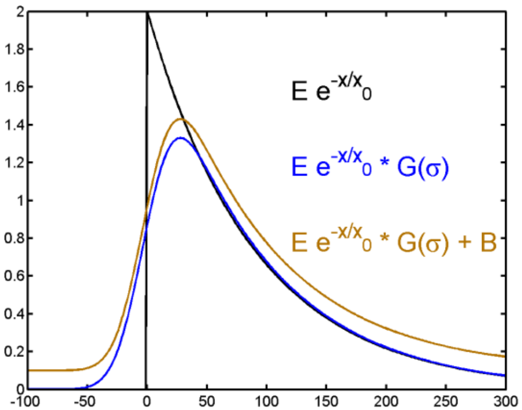


Fit: Truncated exp convolved with Gaussian plus background



Source position may be fitted as well

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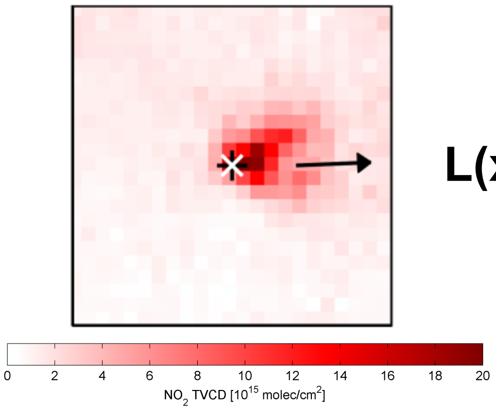

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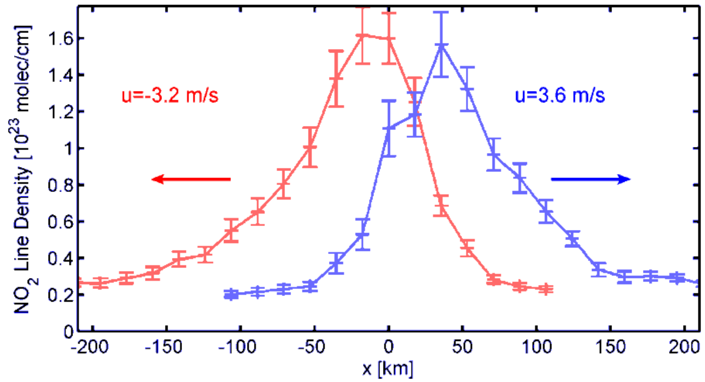

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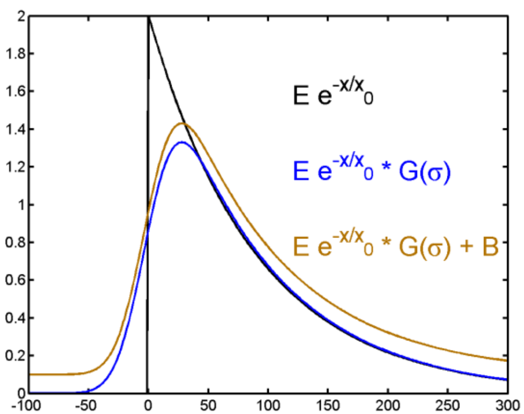

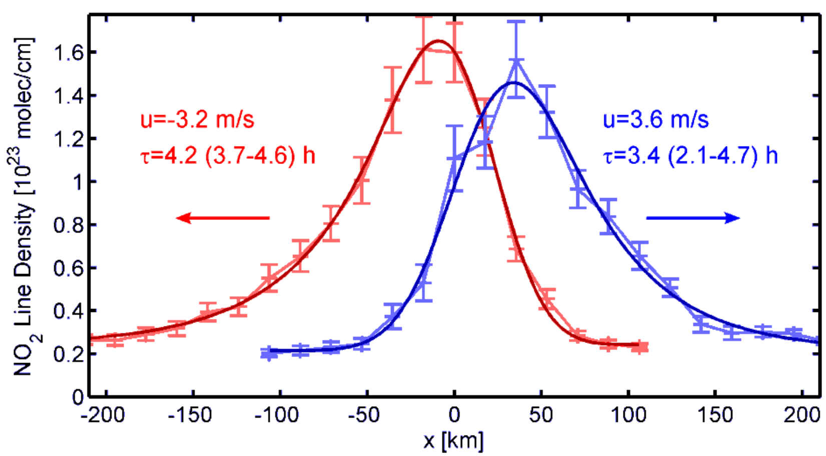

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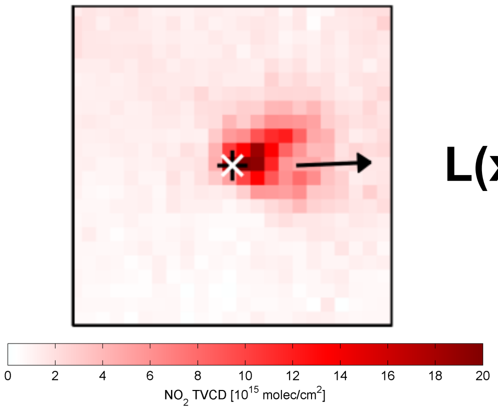
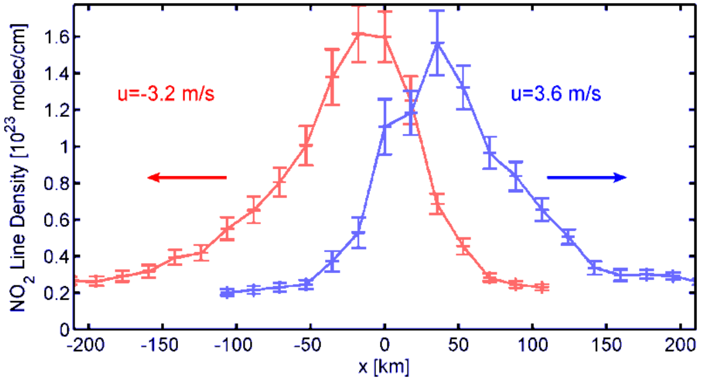


L(x)=∫Vdy



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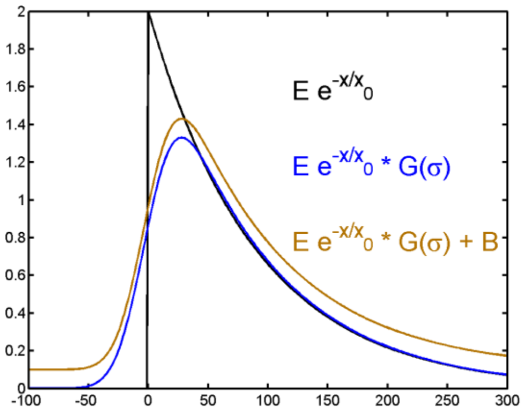

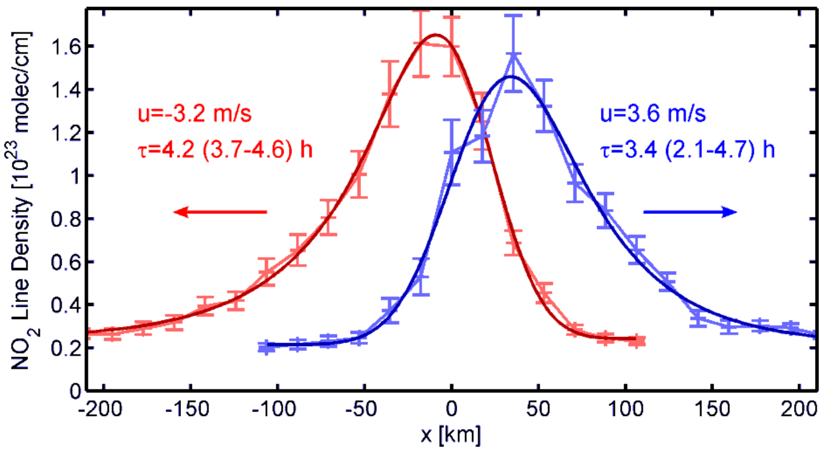

Time constant: $\tau = x_0/w$

E_{NOx}=1.3E

NO_x lifetimes and emissions

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- Downwind decay can be well described by a **single e-folding distance** → **effective lifetime τ_{eff}** - despite **nonlinearities**:

NO_x lifetimes and emissions

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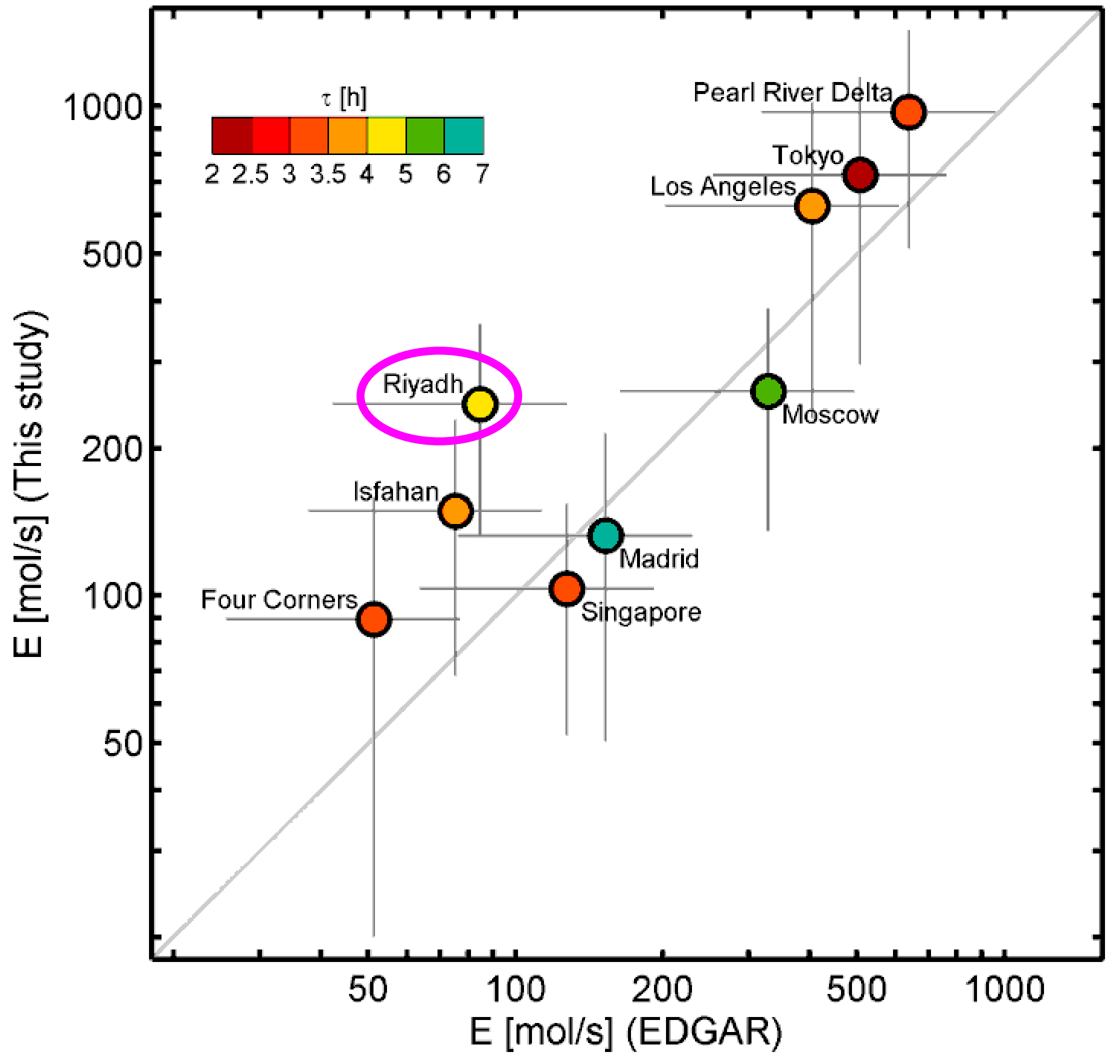
  - Smoothing effects!
- **τ_{eff}** relates **emissions** to **columns**!
- **Riyadh**:

  τ_{eff} =4±0.4 h

  E_{NOx}=246 mol/s

NO_x lifetimes and emissions

- Applicable to strong "point sources" with low background
- E generally in agreement with EDGAR (except Riyadh!)



EDGAR version 4.1. http://edgar.jrc.ec.europa.eu/

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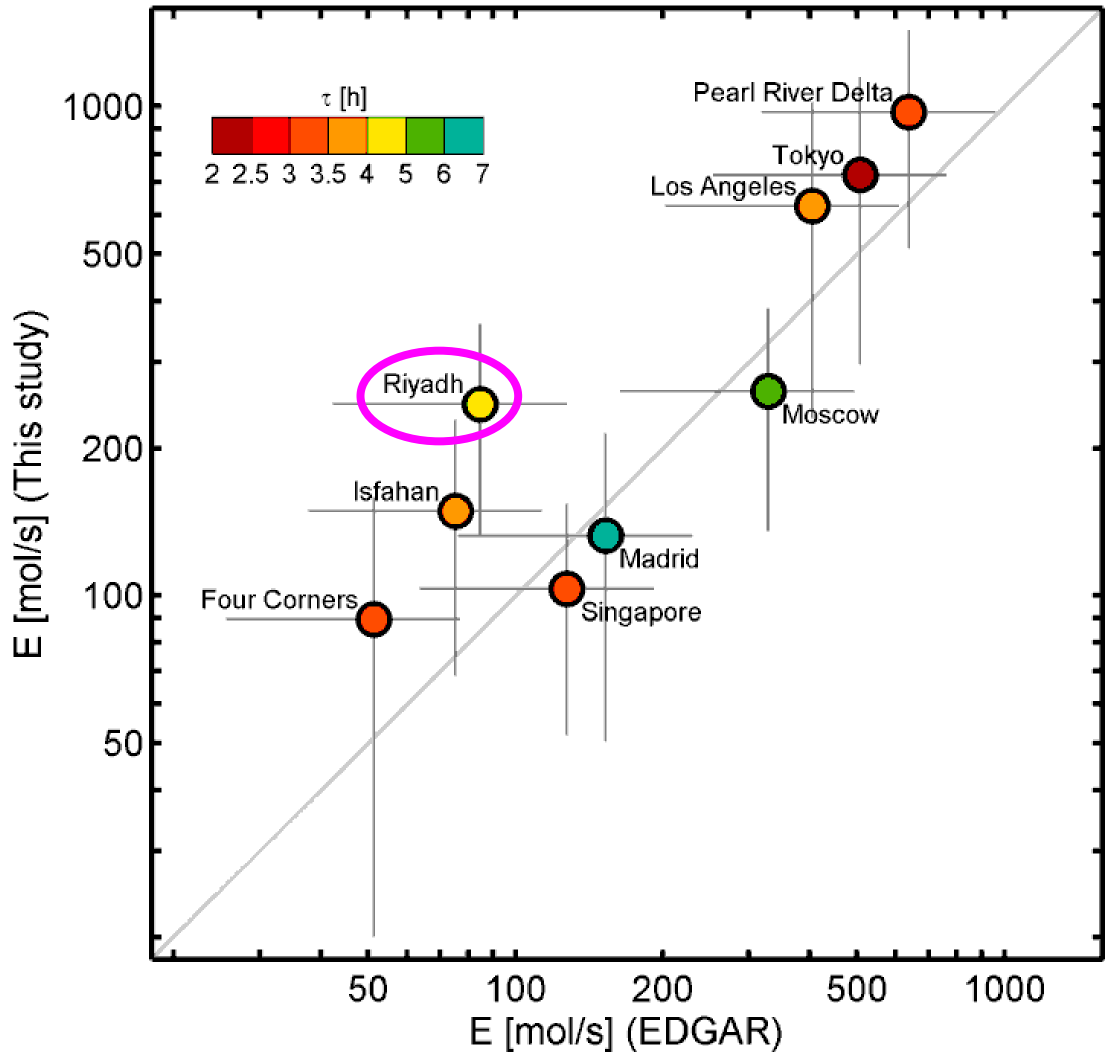
Riyadh (~7M people) is highly polluted! High ozone levels! Should be kept in mind, even if <10M!



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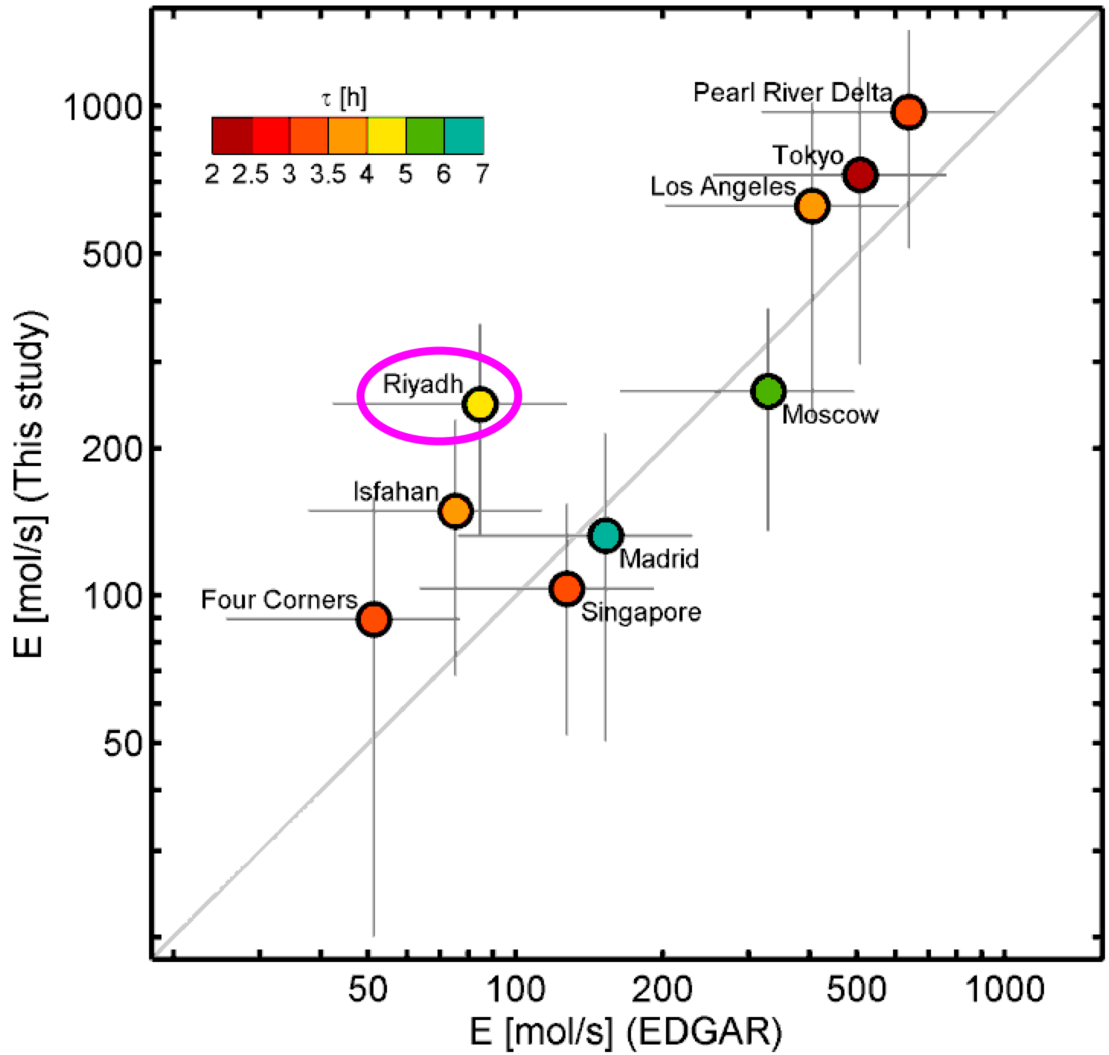

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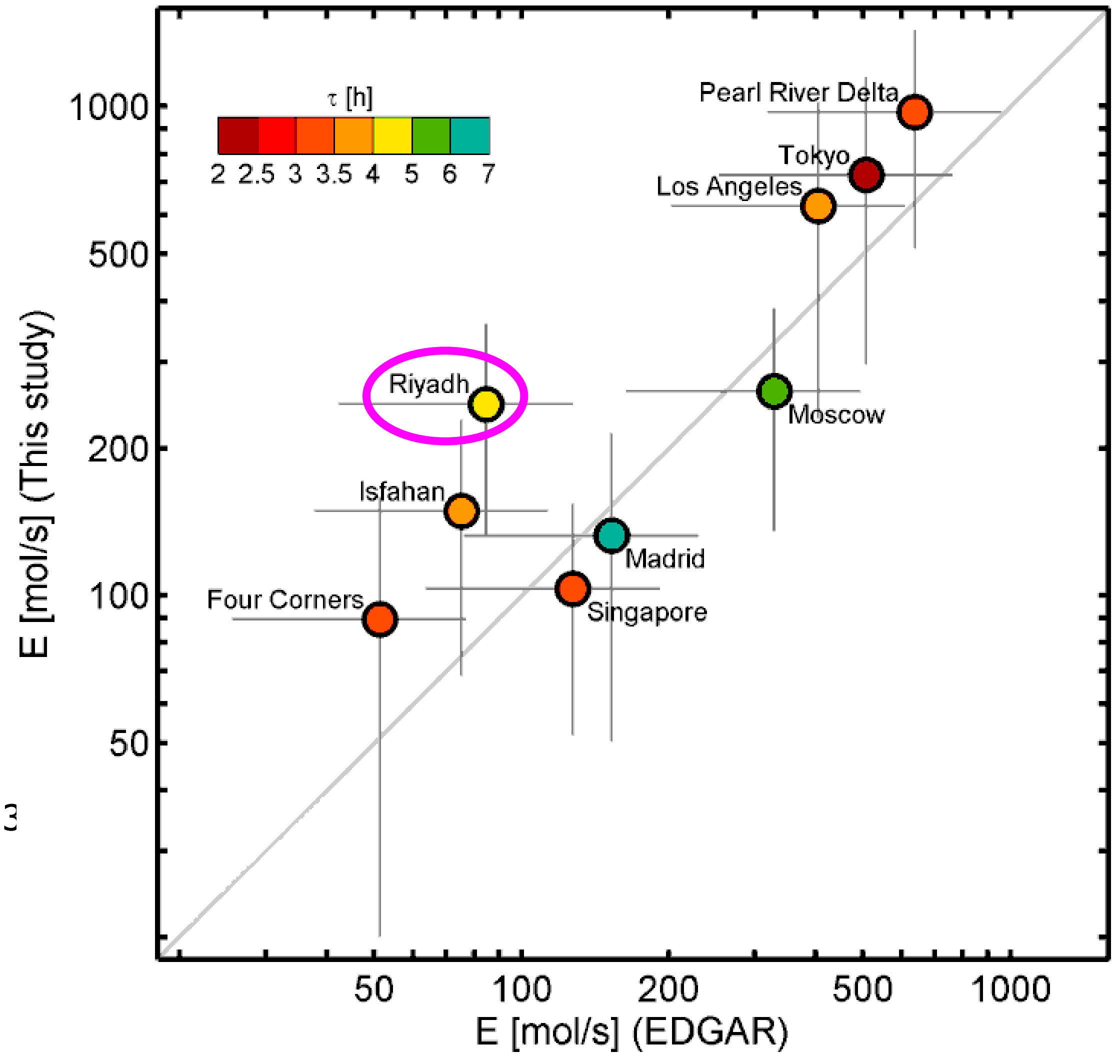
→ OH~4-10 x10^6 molec/cm³



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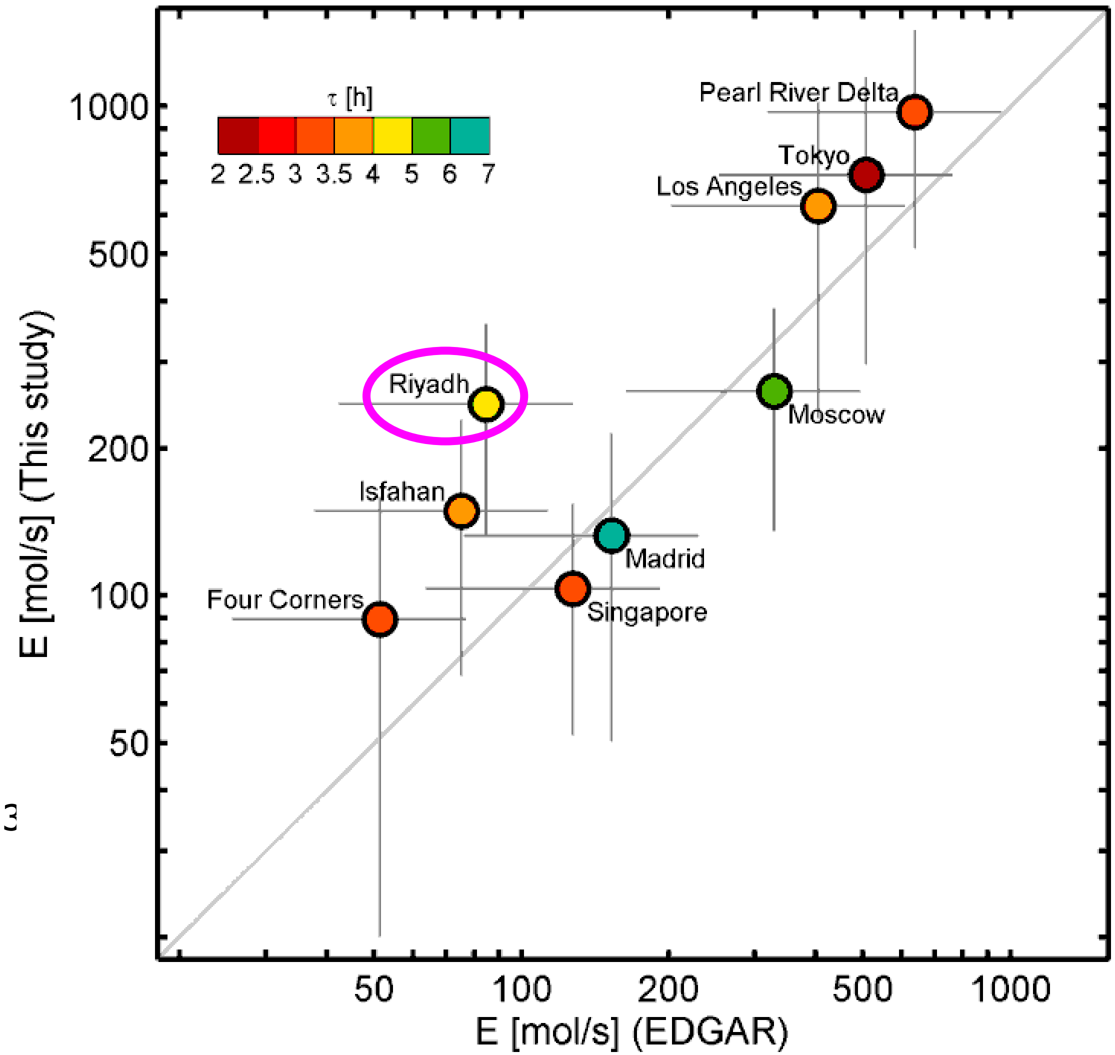
$\rightarrow$ OH~~~4-10~~ x10^6 molec/cm³

6-14



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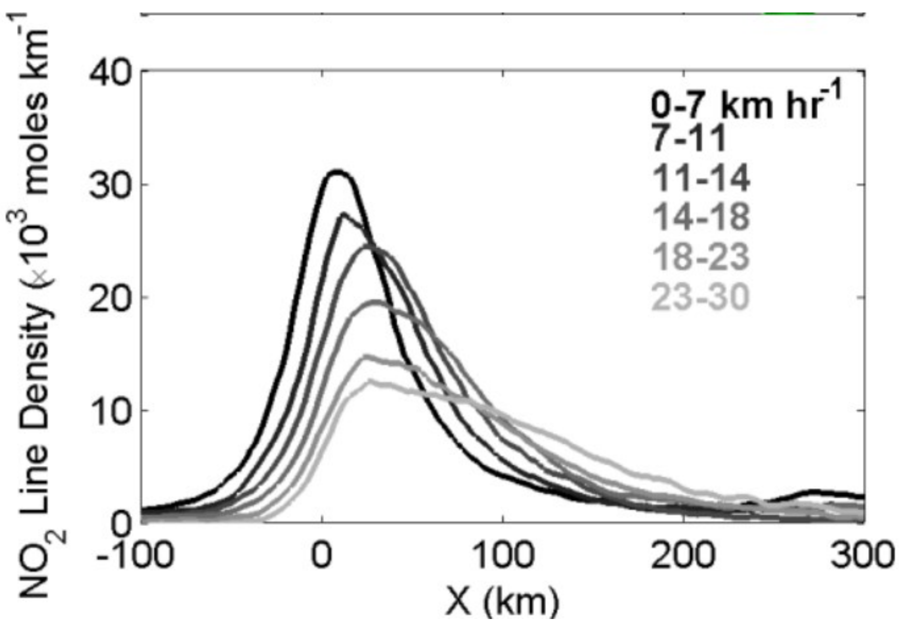
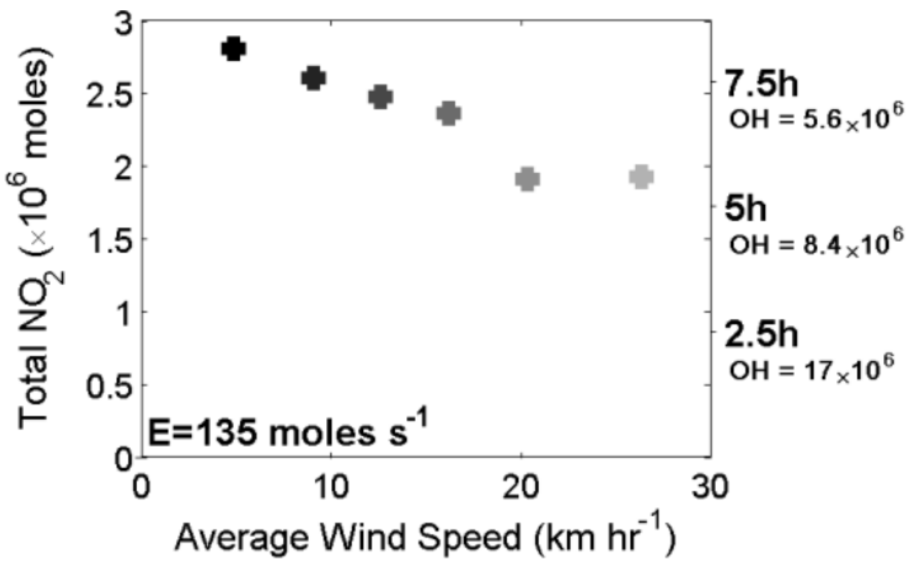
New study: Valin et al., GRL, 2013

- Focus on Riyadh
- Downwind decay for different wind speeds separately
- Results:
  - τ depends on wind speed
  - τ = 6.7 h 4.0 h
  - E_{NOx} = 135 mol/s 246 mol/s



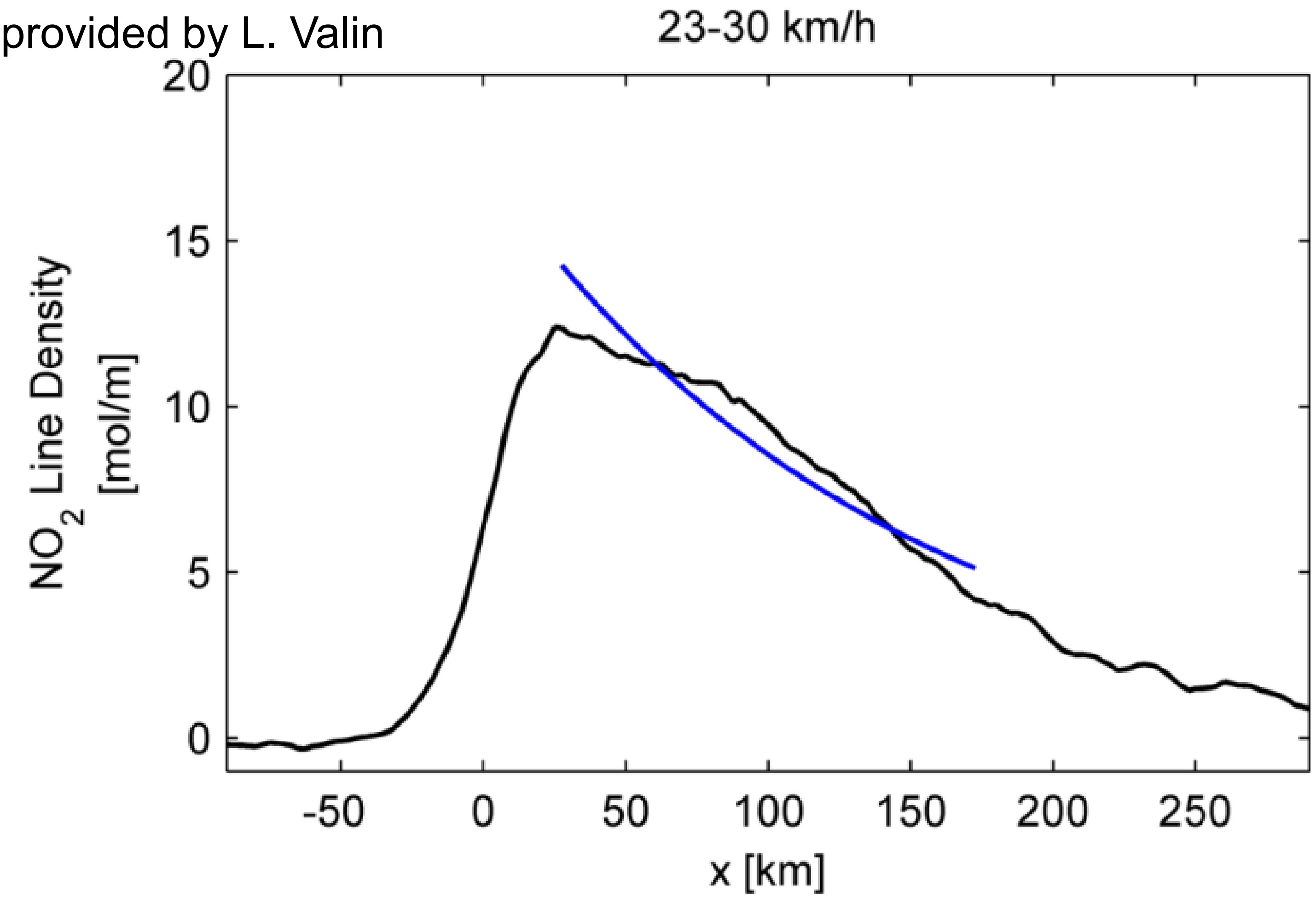


Valin et al., 2013

Valin et al. vs. Beirle et al. vs. Beirle reprocessing

MAX-PLANCK-INSTITUT FÜR CHEMIE

NO_2 LD provided by L. Valin



τ = 5.5 h

E_{NOx} = 135 mol/s (spatial int.)

Valin et al. vs. Beirle et al. vs. Beirle reprocessing

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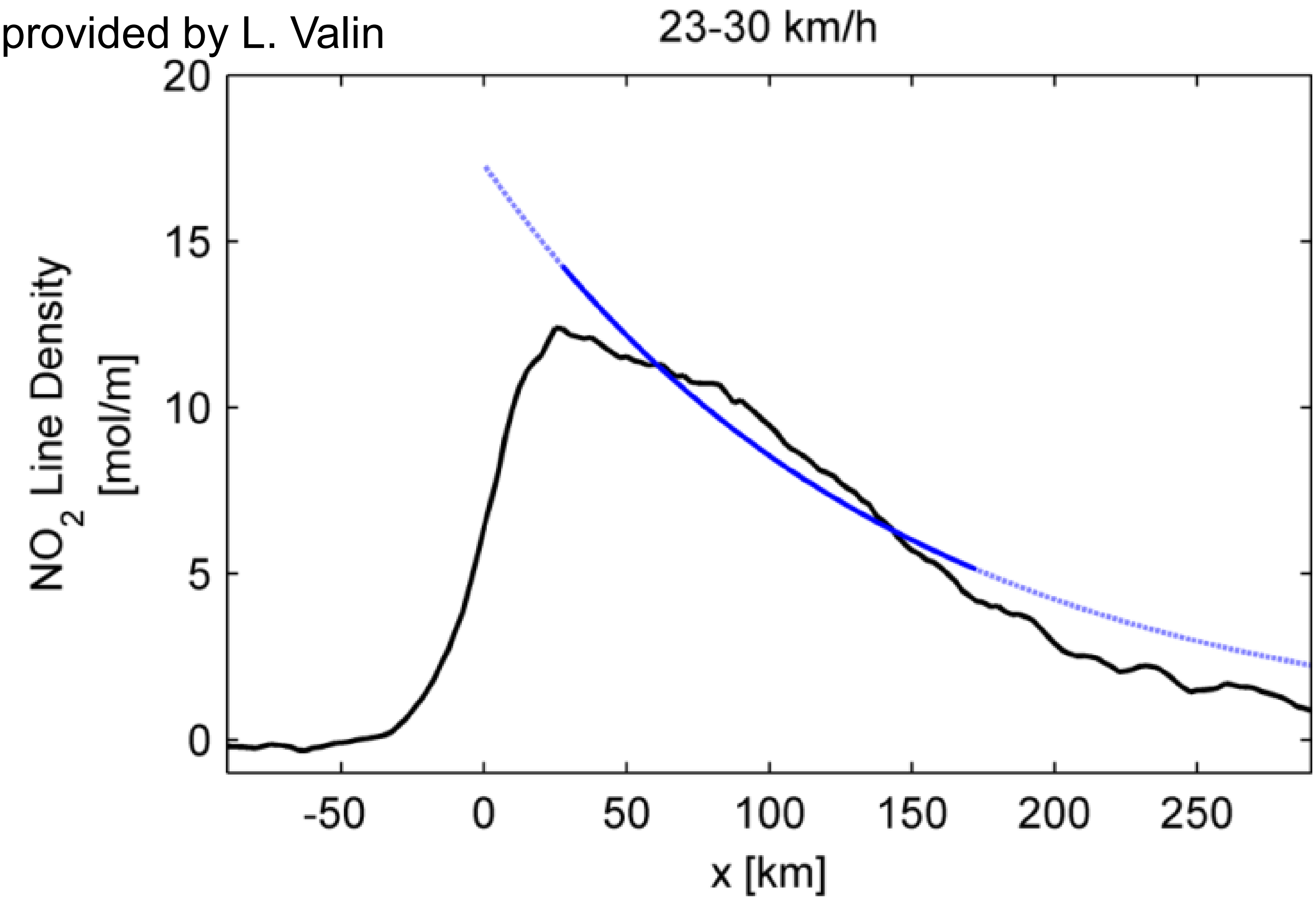

τ	=	5.5	h
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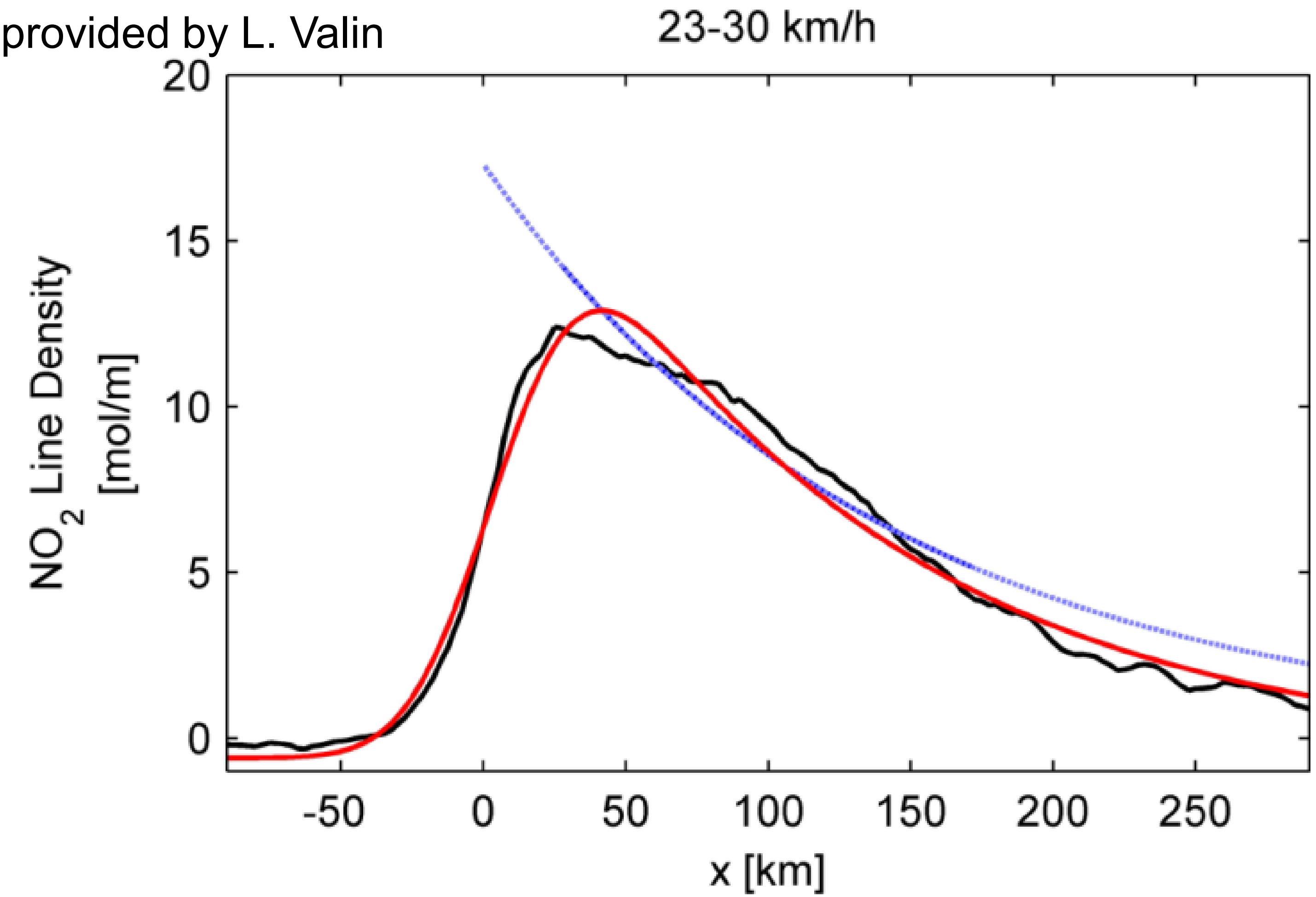
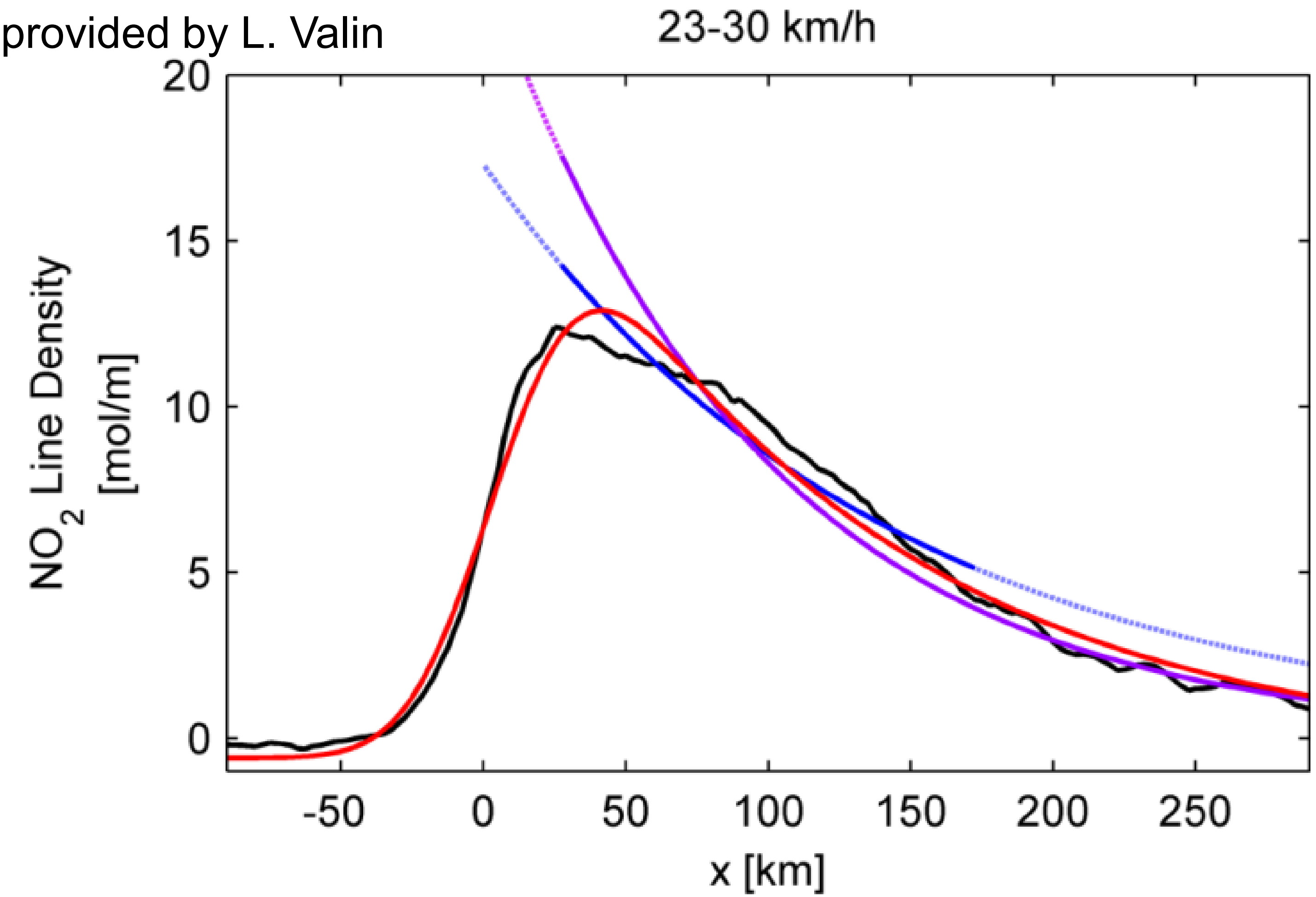


τ	=	5.5	4.5	h
E_{NOx}	=	135	188	mol/s

Valin et al. vs. Beirle et al. vs. Beirle reprocessing

MAX-PLANCK-INSTITUT FÜR CHEMIE

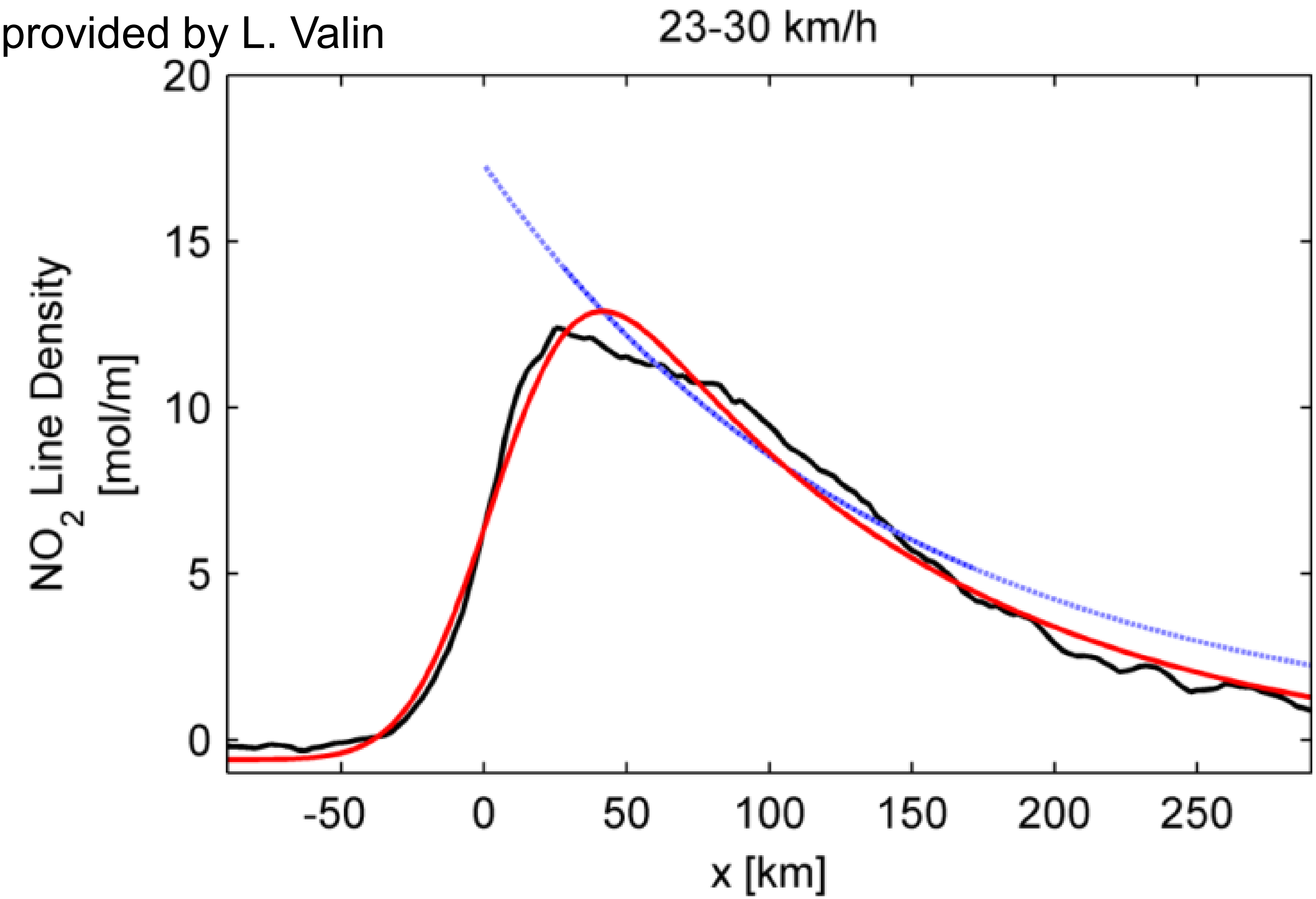
NO_2 LD provided by L. Valin



τ	=	5.5	3.6	4.5	h
E_{NOx}	=	135	191	188	mol/s

Valin et al. vs. Beirle et al. vs. Beirle reprocessing

NO_2 LD provided by L. Valin



τ	=	5.5	4.5	h	-18%
E_{NOx}	=	135	188	mol/s	39%

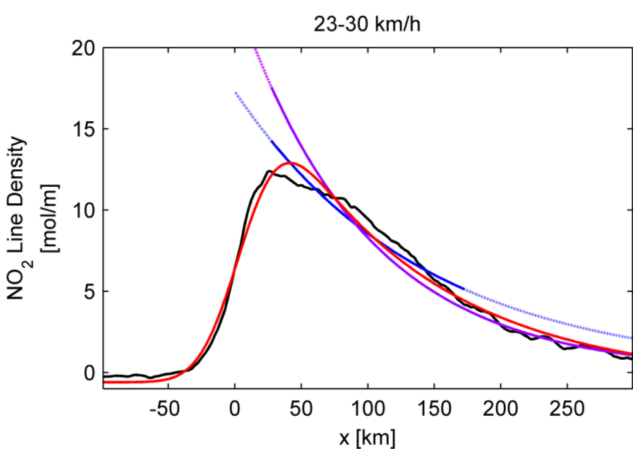
Valin et al. vs. Beirle et al. vs. Beirle reprocessing

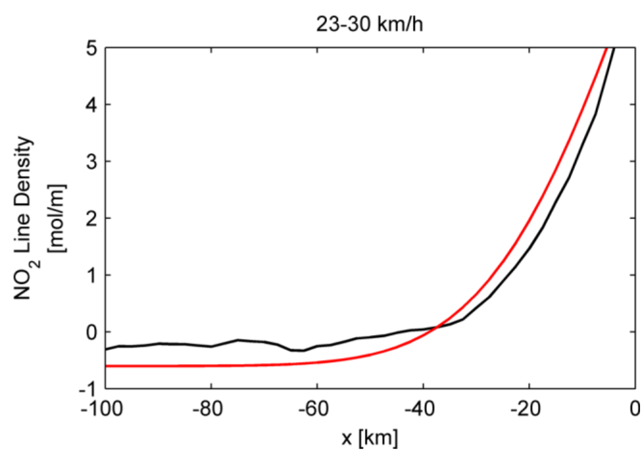
MAX-PLANCK-INSTITUT FÜR CHEMIE

τ	=	5.5	4.5	h	-18%
E_{NOx}	=	135	188	mol/s	39%

E is biased low:

- Integrated column to 300 km (instead of infinity): 8-12%
- Negative Background 7-13%





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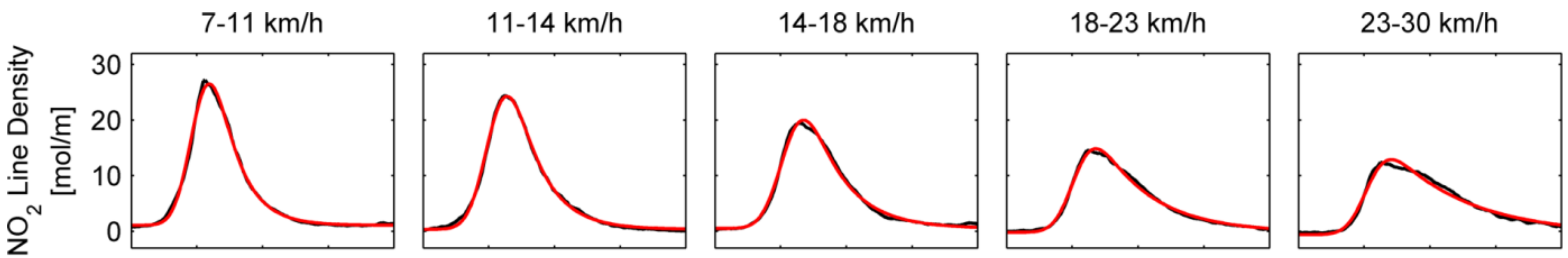

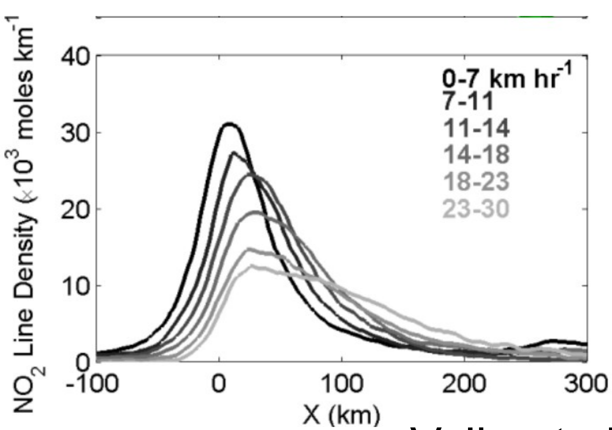

Valin et al., 2013

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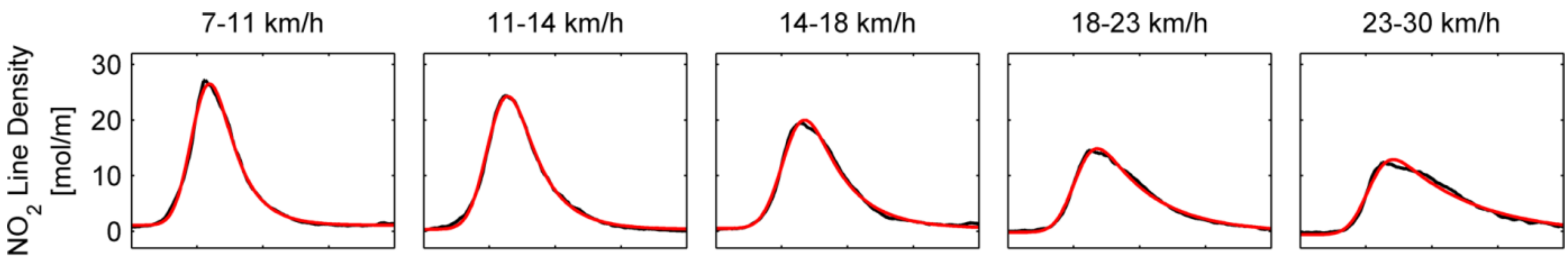

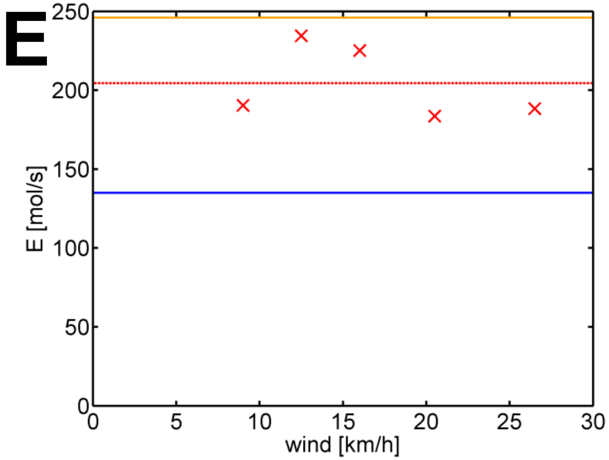

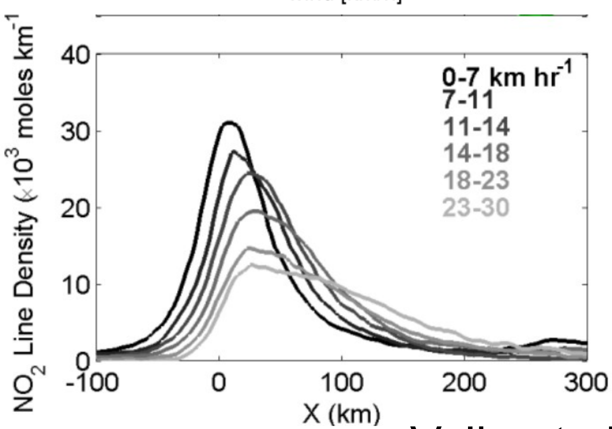

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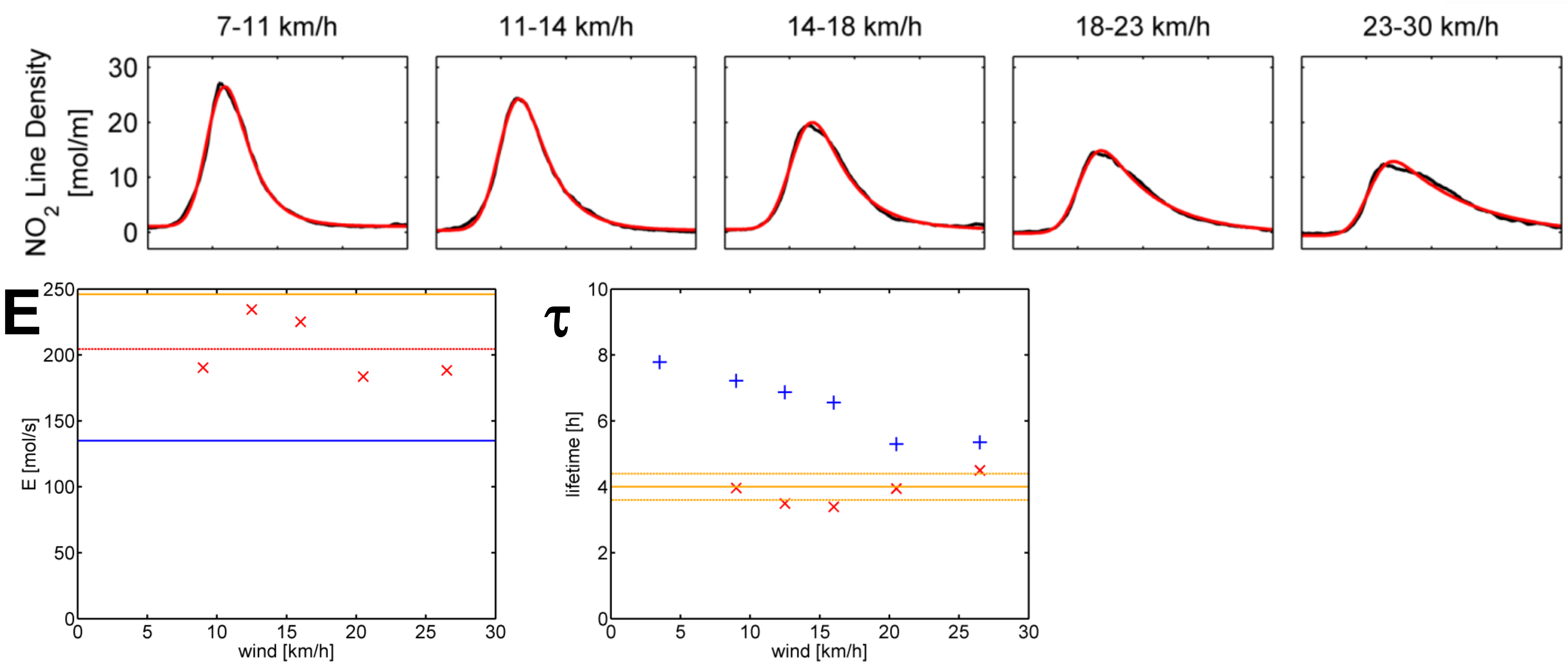

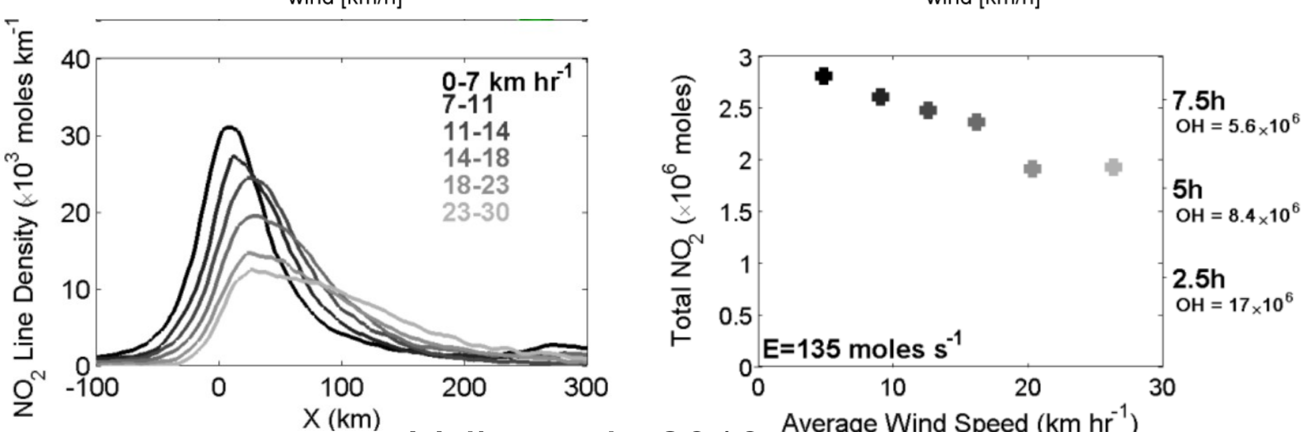

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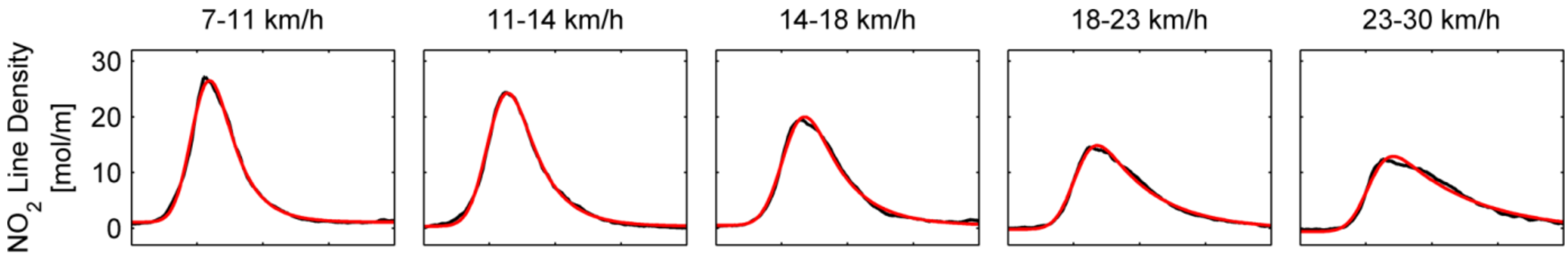

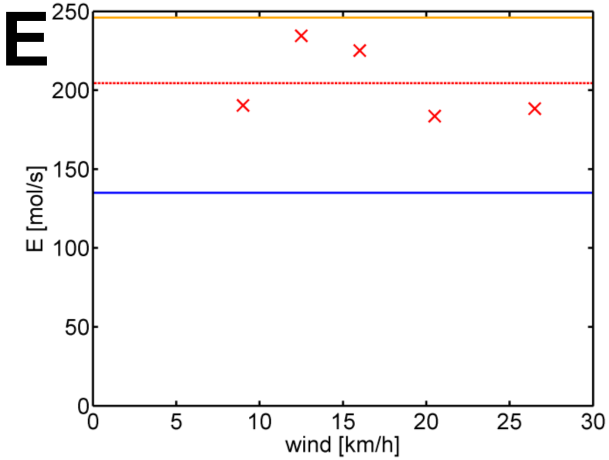

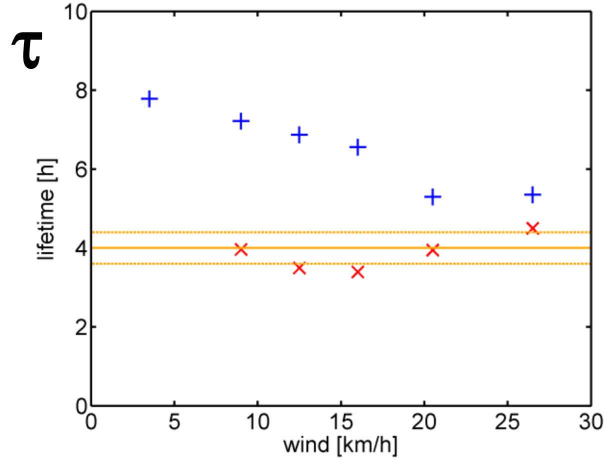

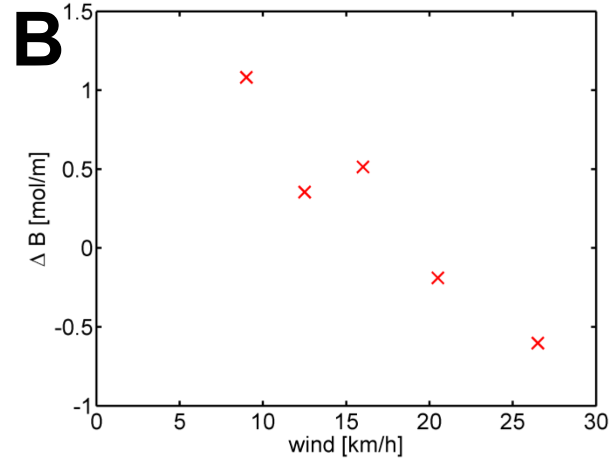

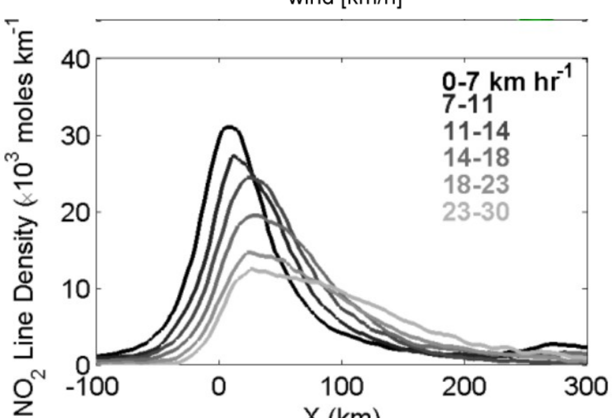

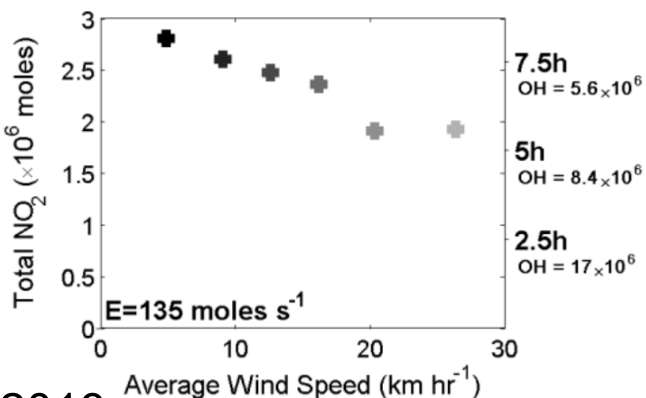

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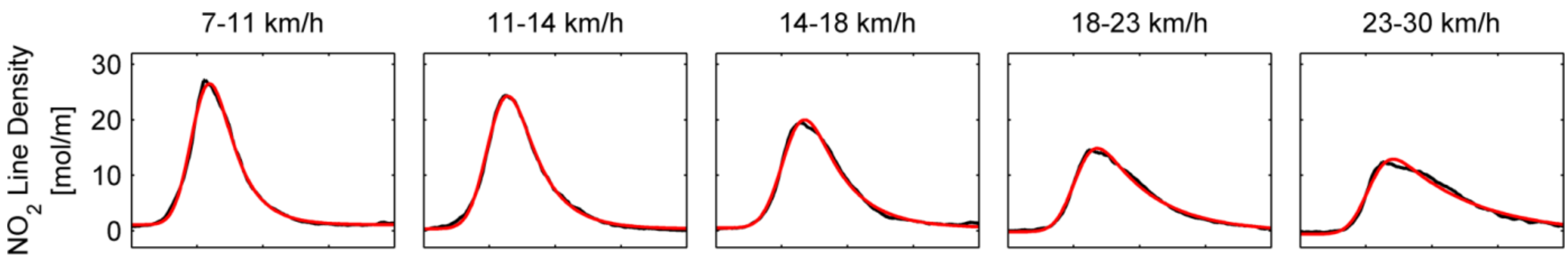
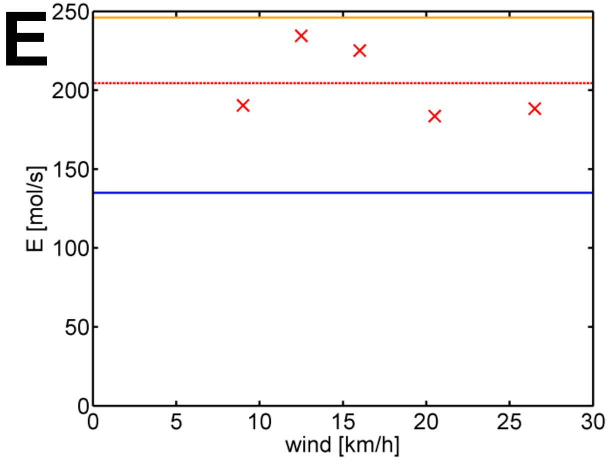
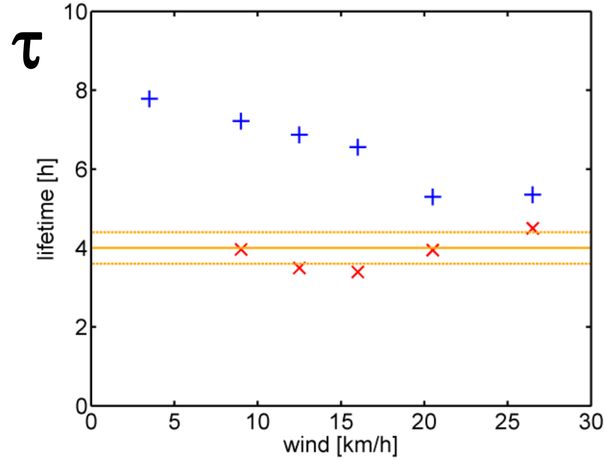
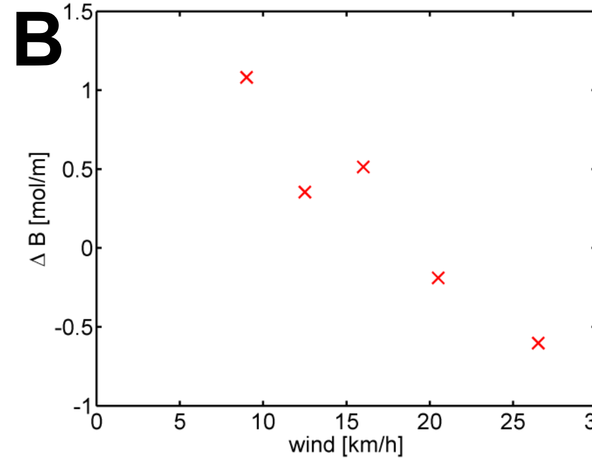
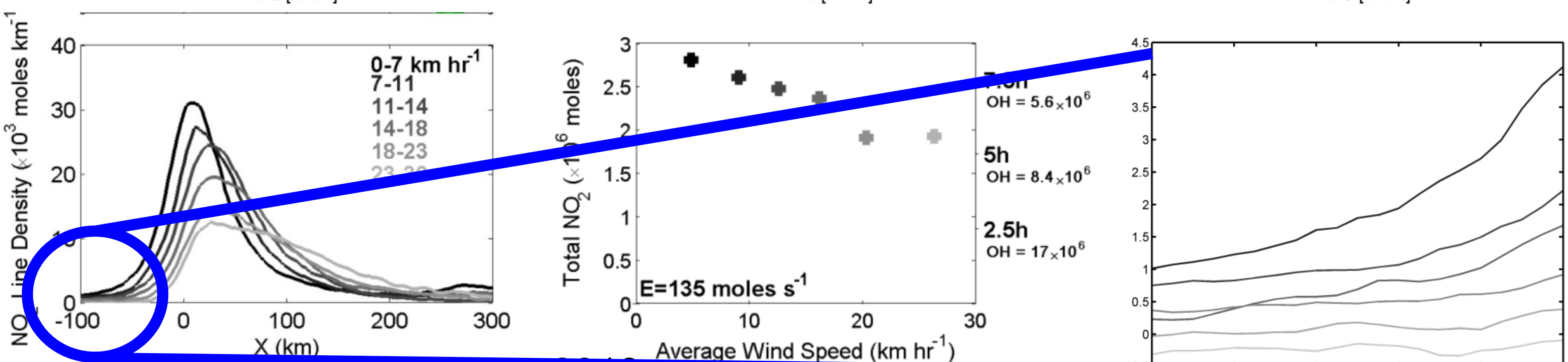
MAX-PLANCK-INSTITUT FÜR CHEMIE





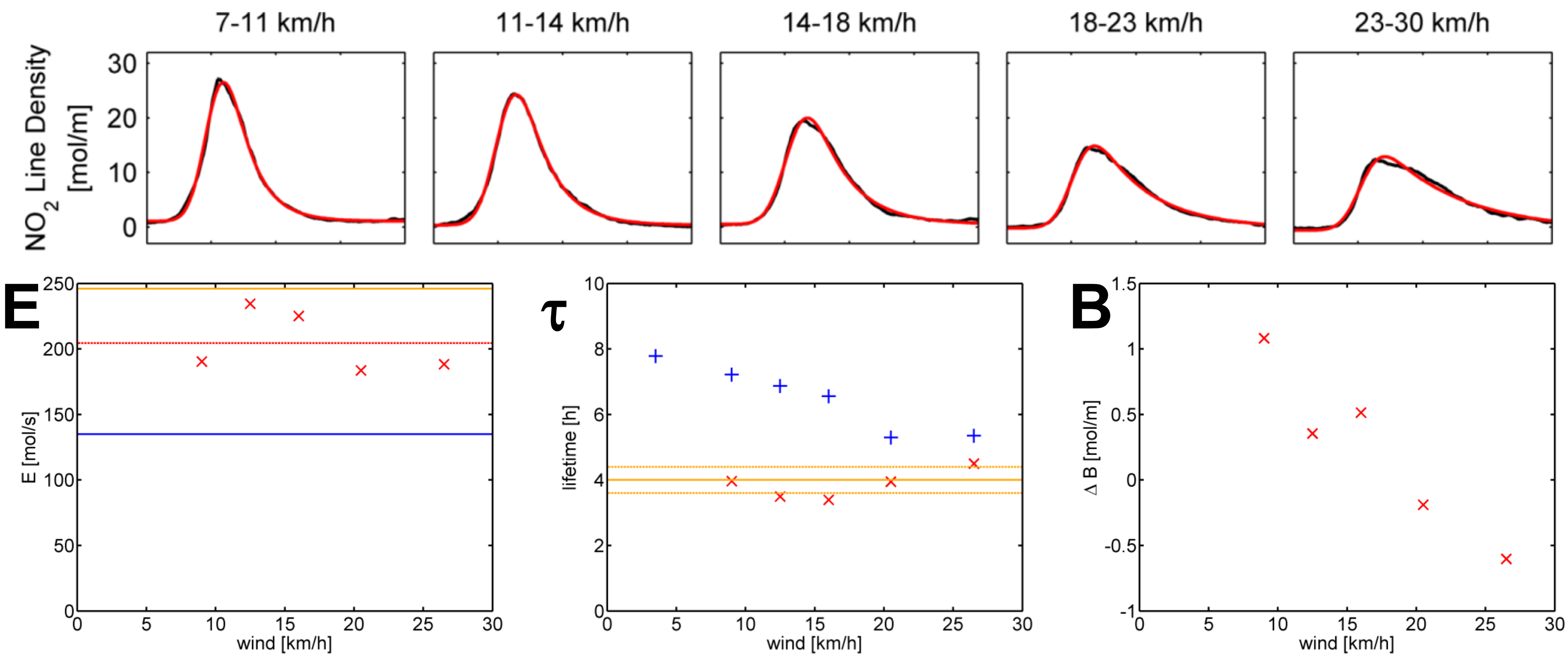


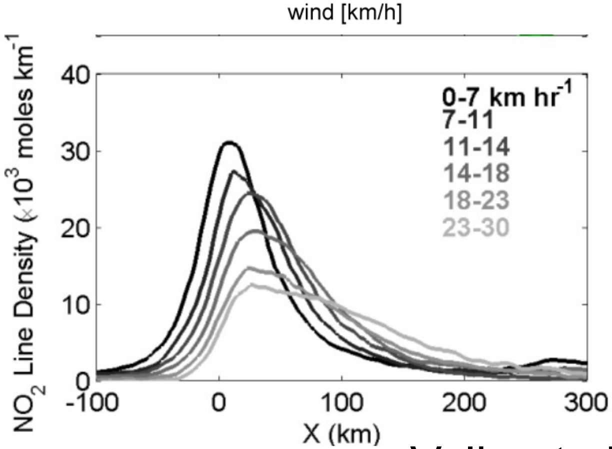


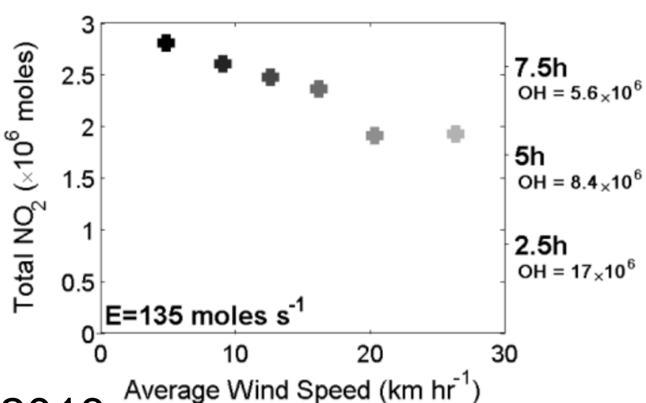


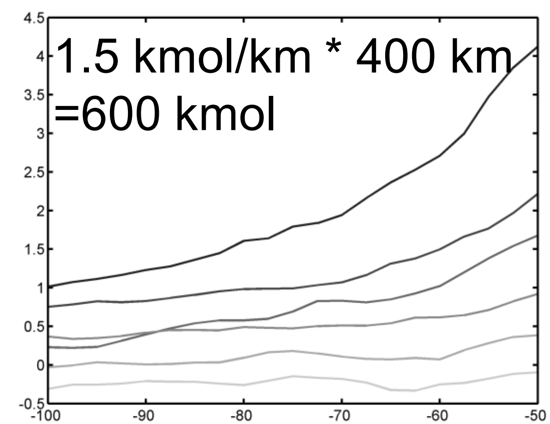
Valin et al., 2013

Valin et al. vs. Beirle et al. vs. Beirle reprocessing









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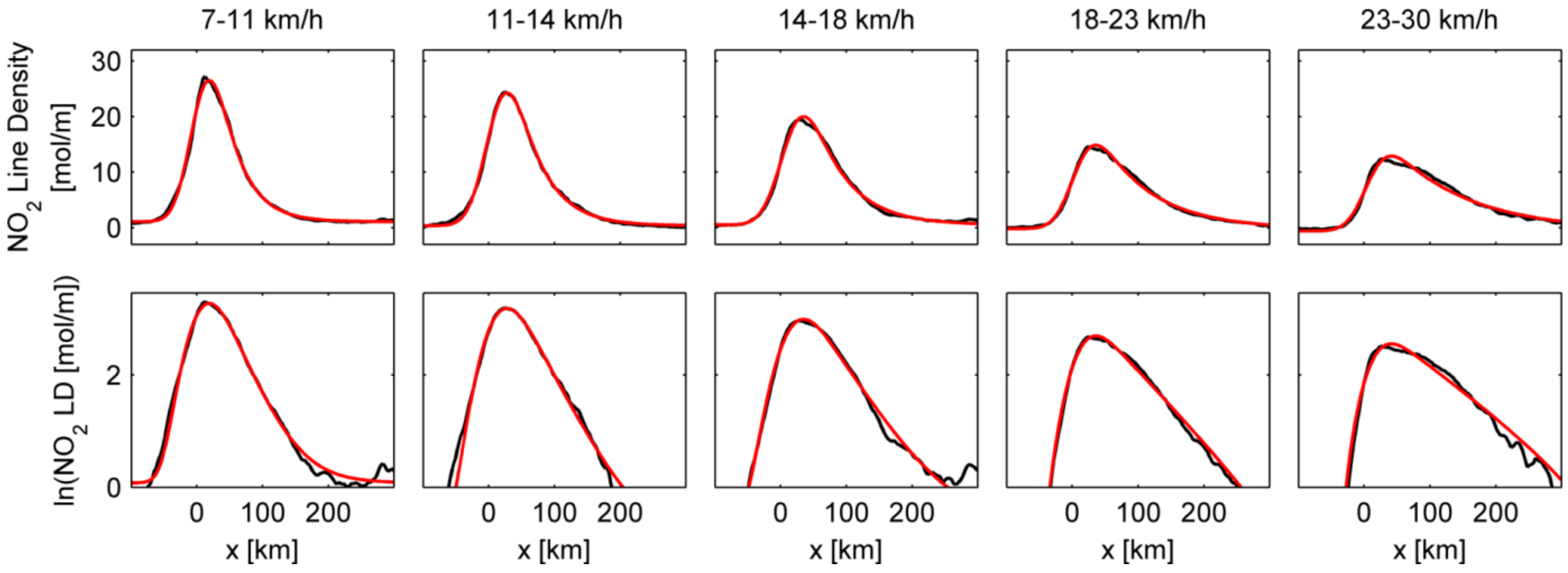

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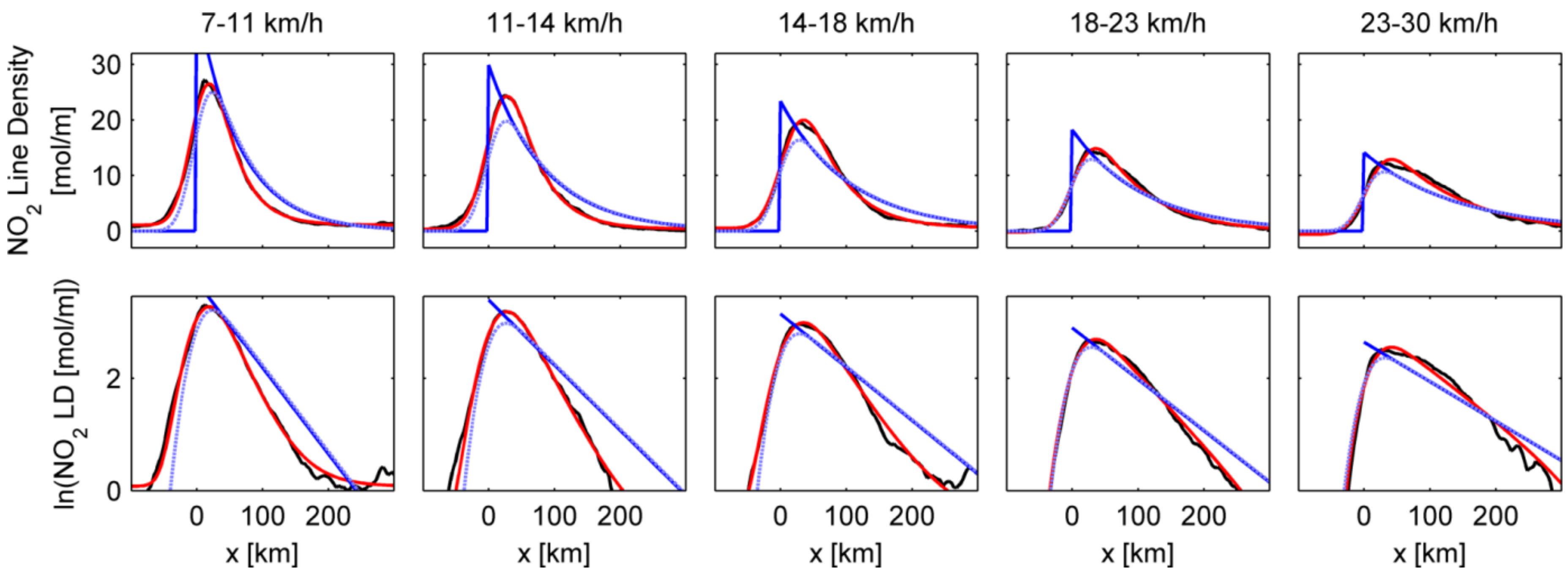
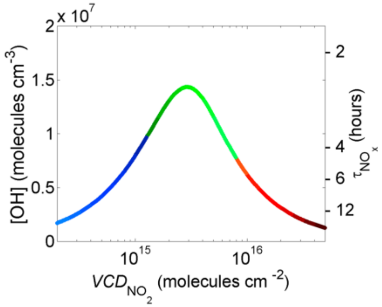


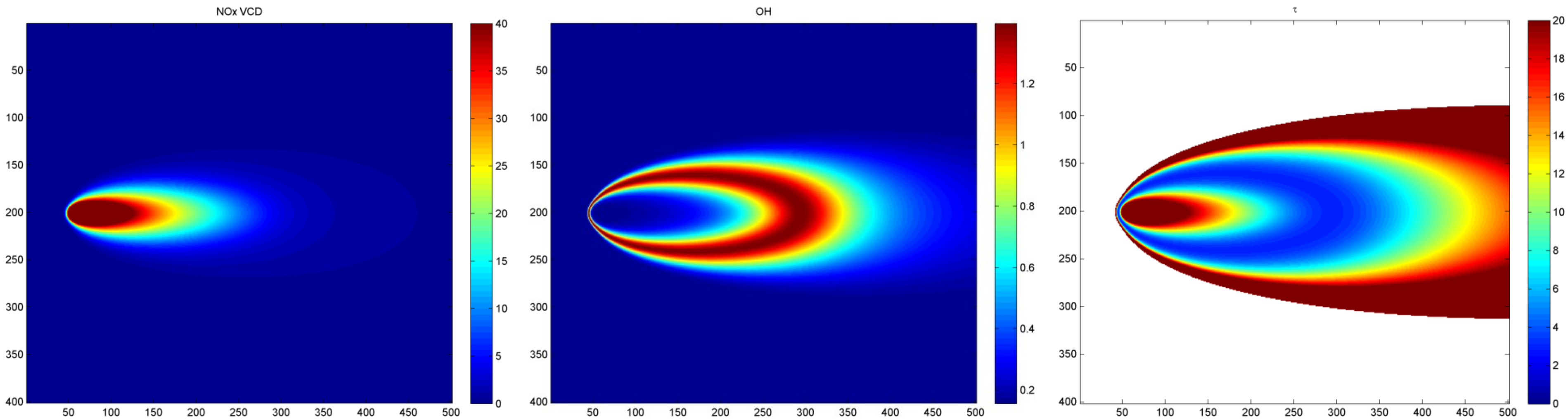
- τ biased high
- $\rightarrow$ E biased low
- No dependency on wind speed !?

Nonlinearities: Downwind change of τ!?

- Simple model: Emissions, transport, dilution, chemistry according to $OH=f(NO_x)$
- Lifetime is
  - High at the source
  - Low at ~ 100-200 km
  - High beyond



Valin et al., 2013



VCD OH τ

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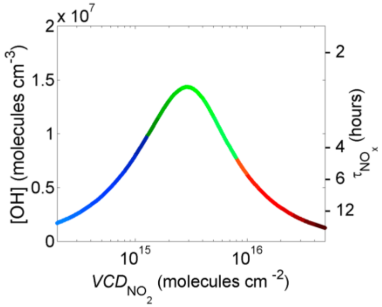
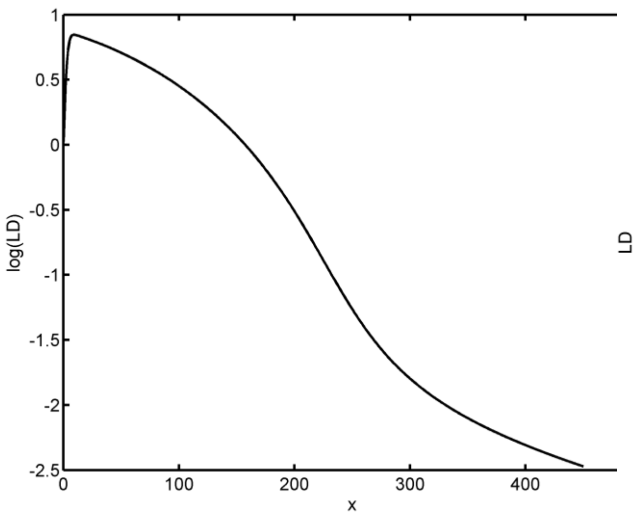
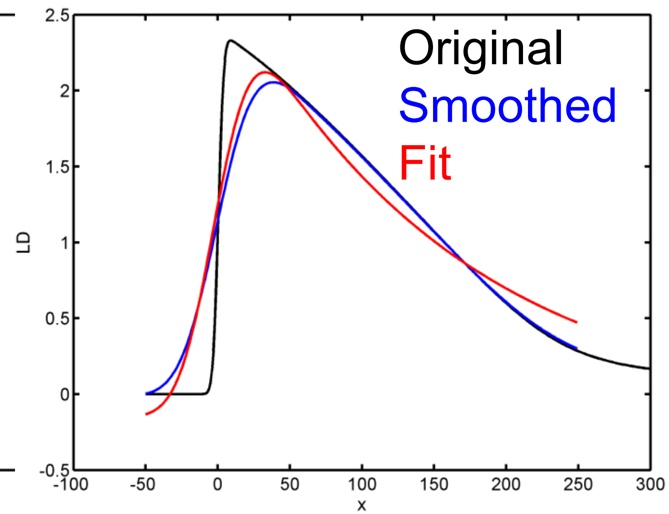


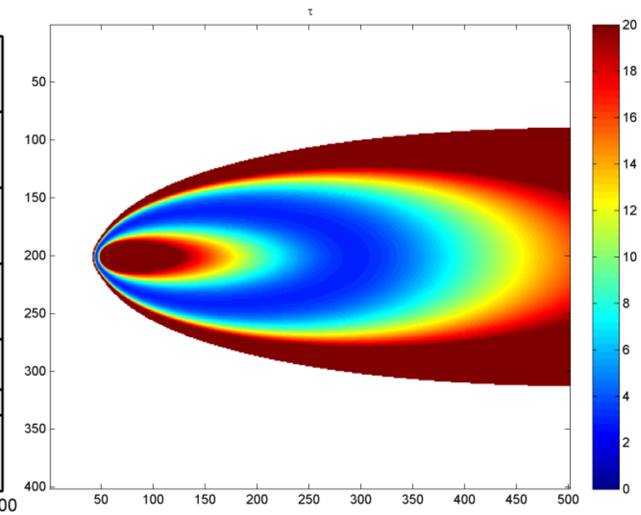
Valin et al., 2013



Log(LD)



LD



τ

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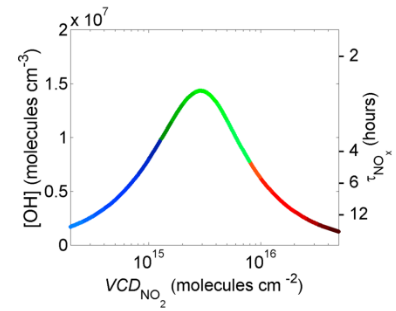
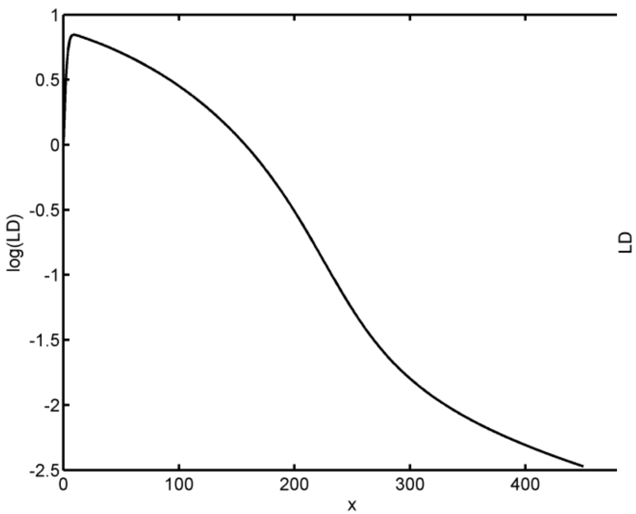


Valin et al., 2013



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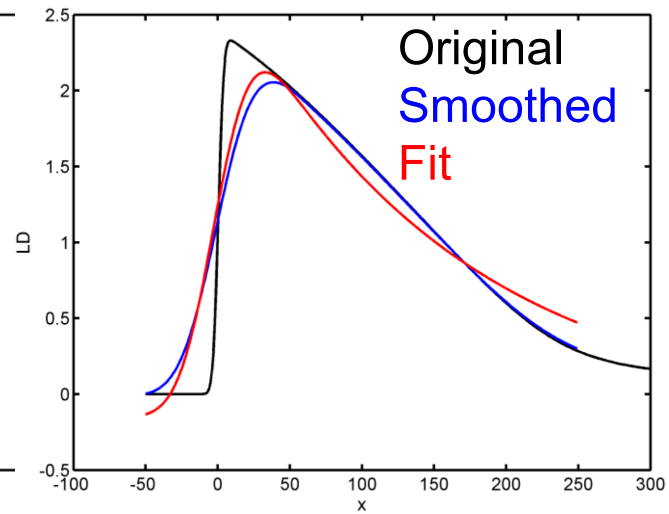
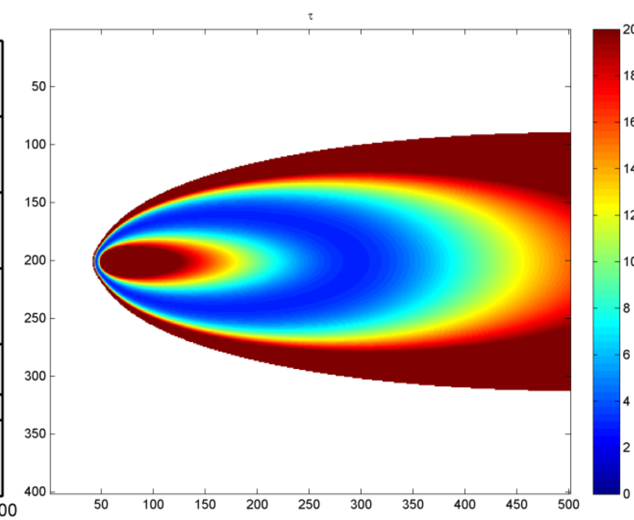


LD



τ

convex or **concave**?

Nonlinearities: Downwind change of τ!?

MAX-PLANCK-INSTITUT FÜR CHEMIE

- Do we see this effect? Not in Beirle et al., 2011! But...

Nonlinearities: Downwind change of τ!?

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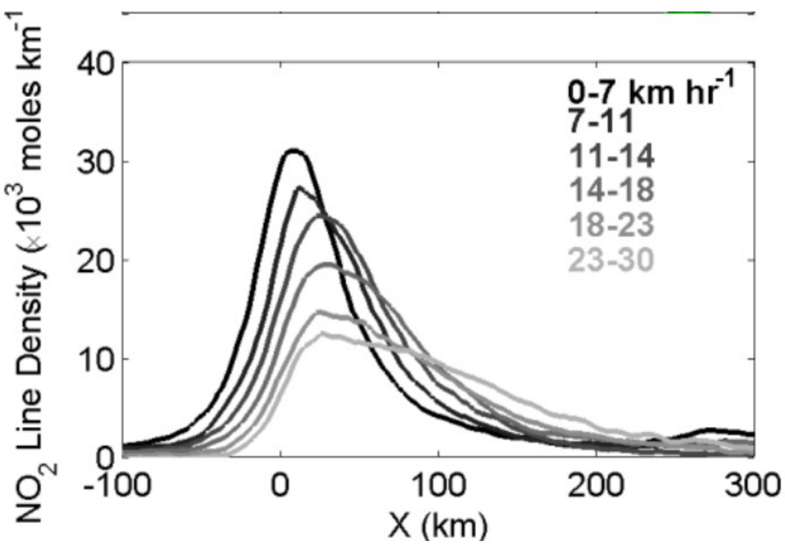
For strong winds?



Valin et al., 2013

Nonlinearities: Downwind change of τ!?

MAX-PLANCK-INSTITUT FÜR CHEMIE

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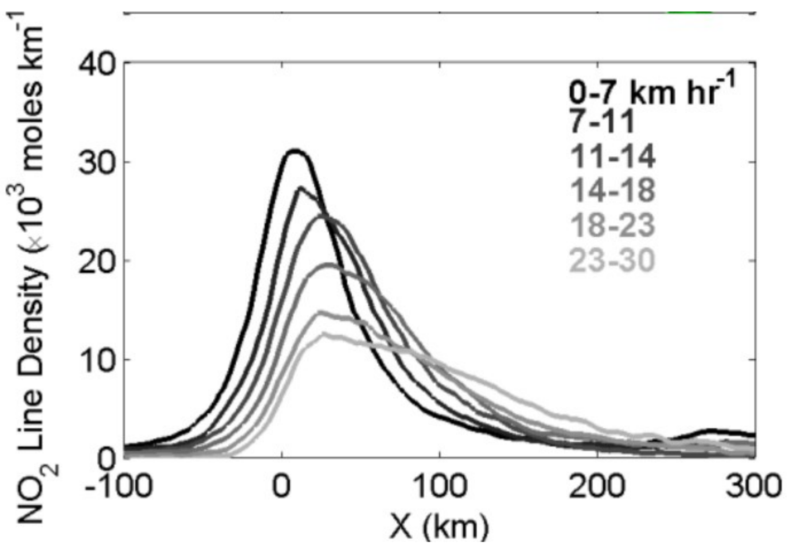
In the shiptracks!?



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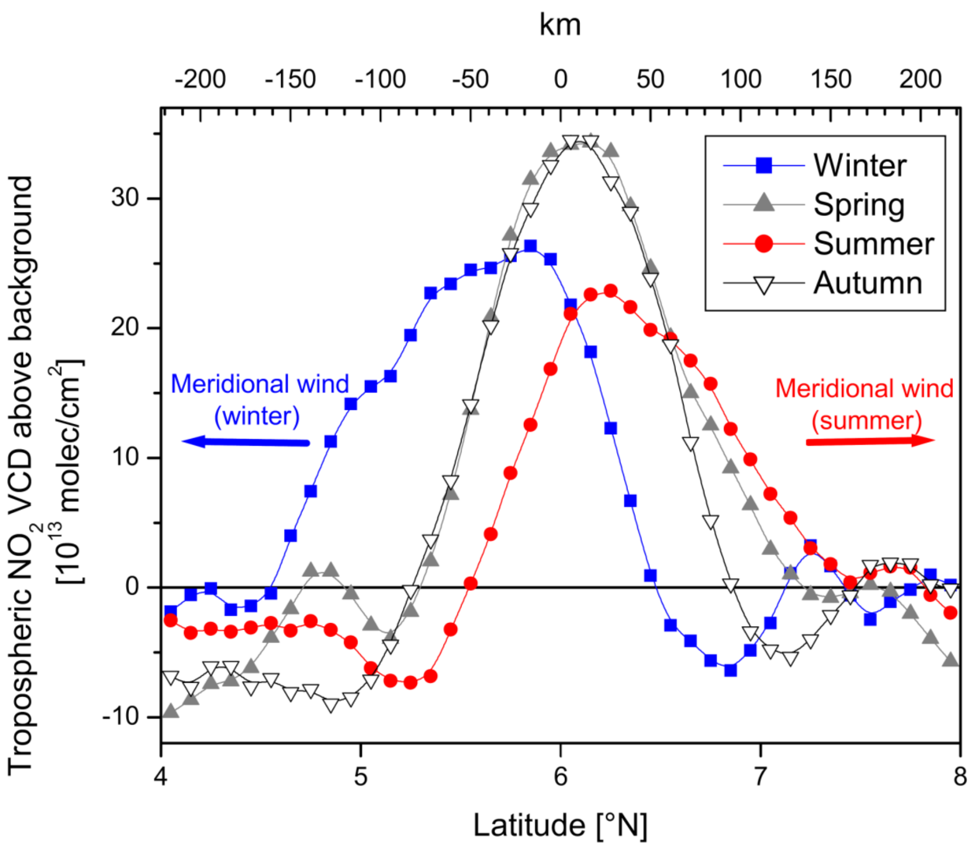

Nonlinearities: Downwind change of τ!?

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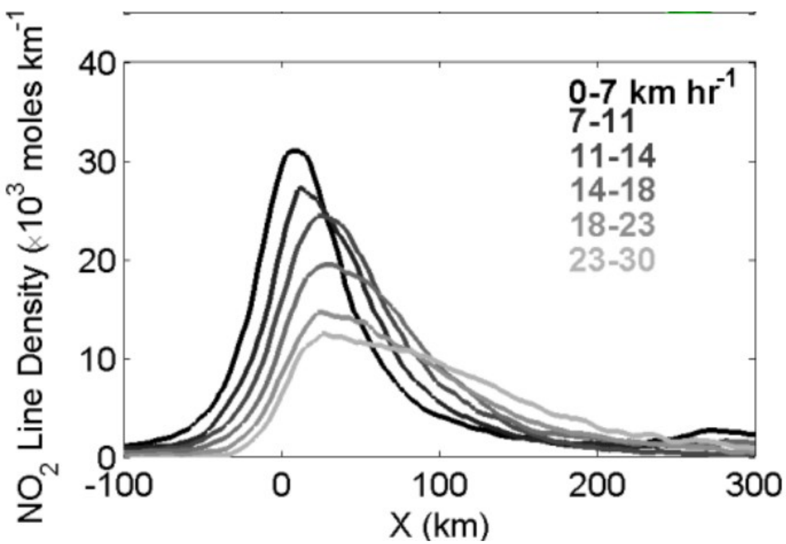
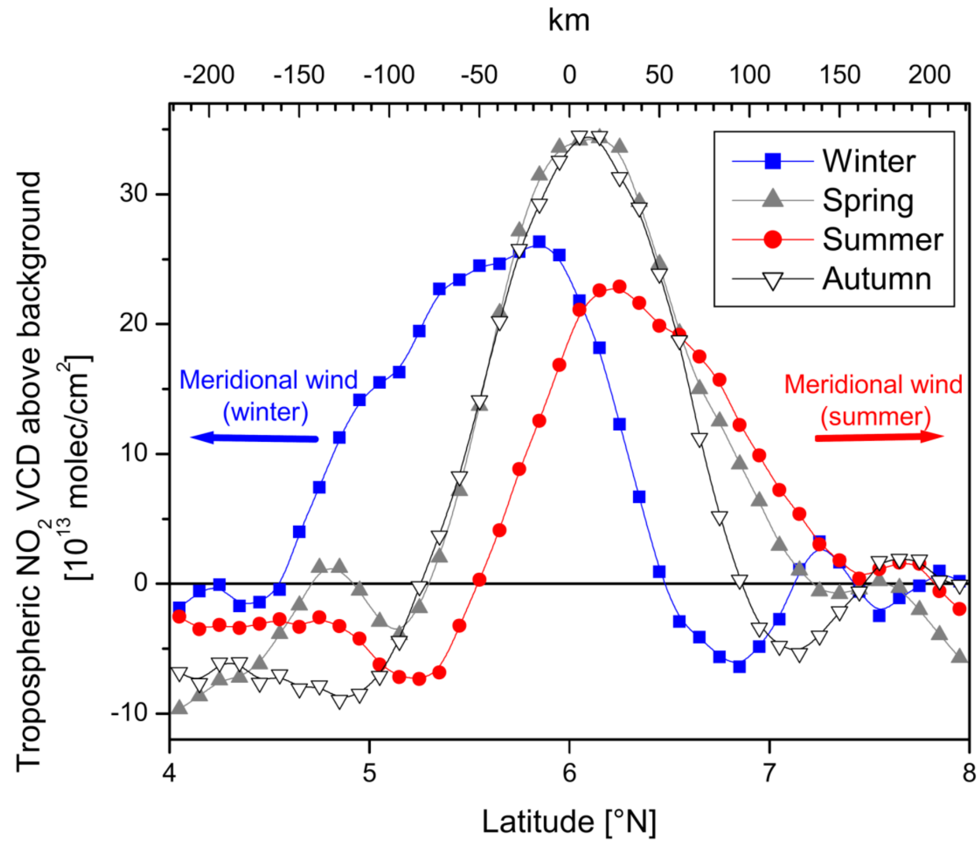


Valin et al., 2013

In the shiptracks!?



- OH is not everything
- Needs further investigation...

Conclusions I: Current status

MAX-PLANCK-INSTITUT FÜR CHEMIE

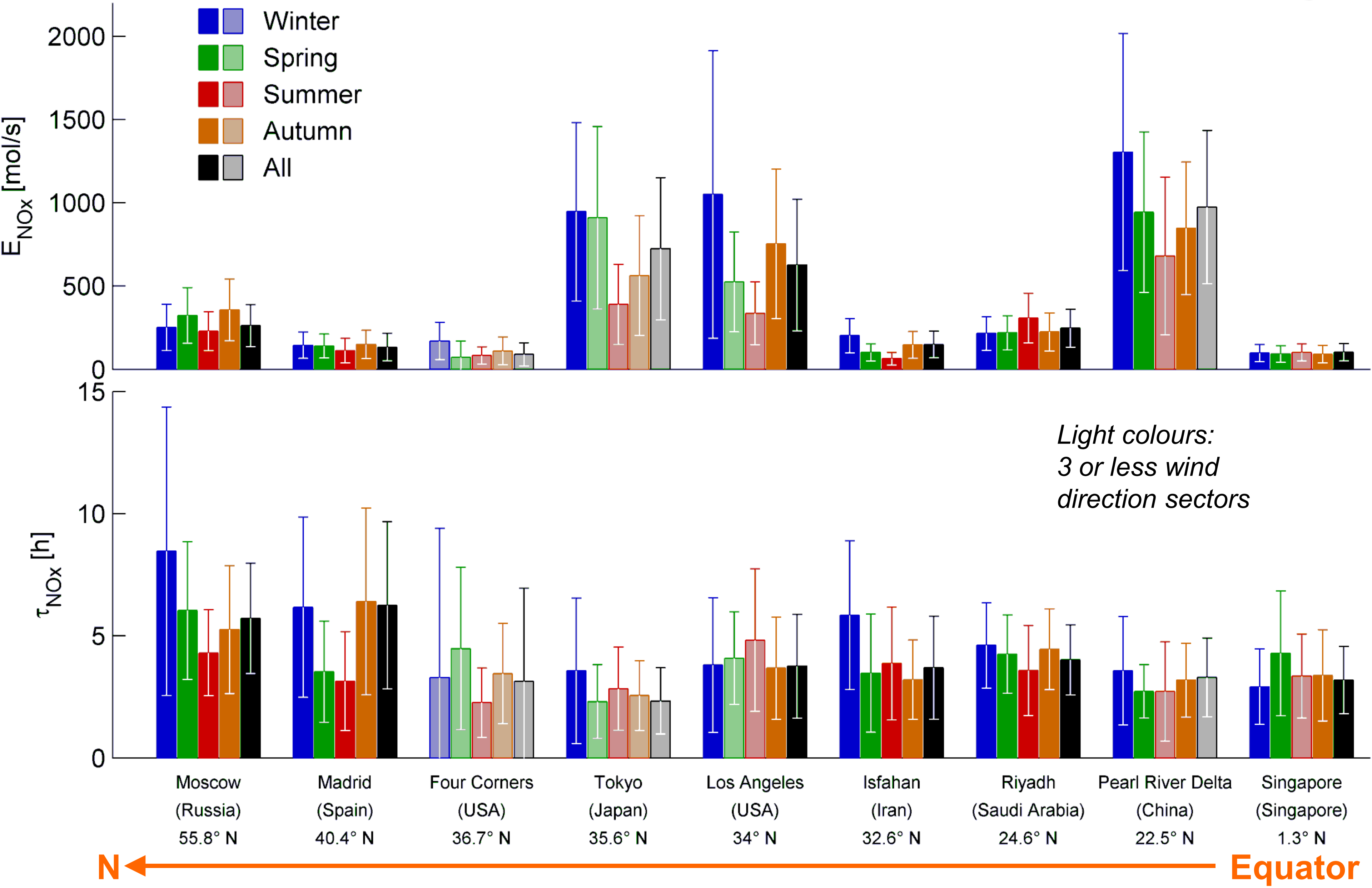
- Satellite measurements provide information about emissions **and chemistry**
- Downwind decay can be described by single **effective** daytime NO_x **lifetime**: ~4h – despite nonlinearities!
- **Smoothing** effects (source distribution, satellite pixel size, dilution) and **Background** have to be considered
- Is there a systematic **in-plume variation**?
  - High wind speeds!?
  - Ship tracks!?
  - Needs further investigation

Conclusions II: Outlook

MAX-PLANCK-INSTITUT FÜR CHEMIE

- Crucial **a-priori**: (relative) NO_2 profile:
  - AMF
  - Wind fields
  - → Profile measurements!
- **LEO:**
  - Analysis of long-term means
  - → Ongoing timeseries with good spatial coverage!
- **Smoothing:**
  - → Better spatial resolution!
- **GEO:**
  - Diurnal cycles!?
  - Temporal plume evolution

Additional Slides



Additional slides

Fit

We perform a non-linear least-squares fit of a simple model function $M(x)$ to the observed NO_2 line densities, as function of the distance x. $M(x)$ is composed of

a) a truncated exponential function

$$e(x) := \exp\left(-(x-X)/x_0\right) \quad \text{for } x \geq X \text{ (downwind), and}$$

$$e(x) := 0 \quad \text{for } x < X \text{ (upwind),}$$

where X is the location of the apparent source (relative to the city center), and x_0 is the e-folding distance downwind,

b) a convolution with a Gaussian function

$$G(x) := \frac{1}{\sqrt{2\pi}\cdot\sigma}\cdot\exp\left(-x^2/2\sigma^2\right)$$

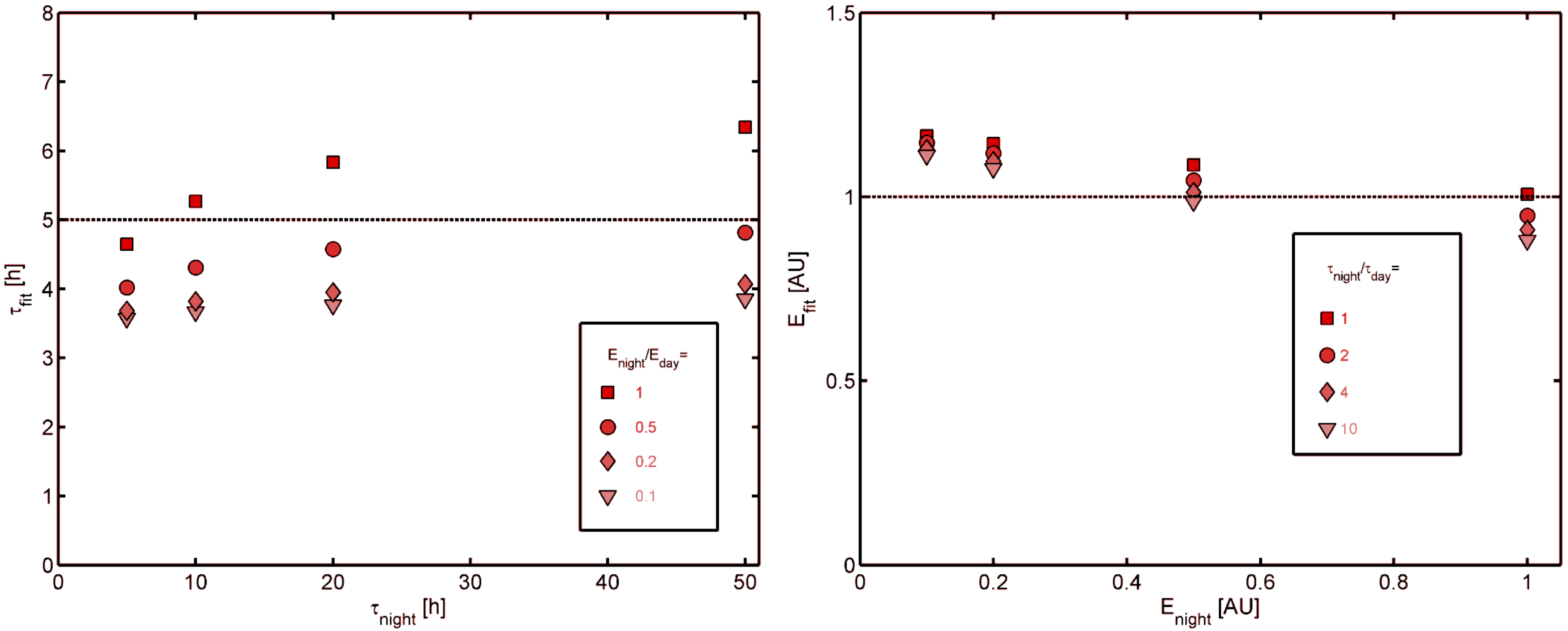
with standard deviation (std) σ, and

c) a scaling by total emissions E, and a constant background B, so that in total

$$M(x) := E\cdot(e\otimes G)(x) + B$$

The fit considers the x-range from 100 km upwind to 200 km downwind. Fitted parameters are x_0, σ, X, E, and B.

Additional slides



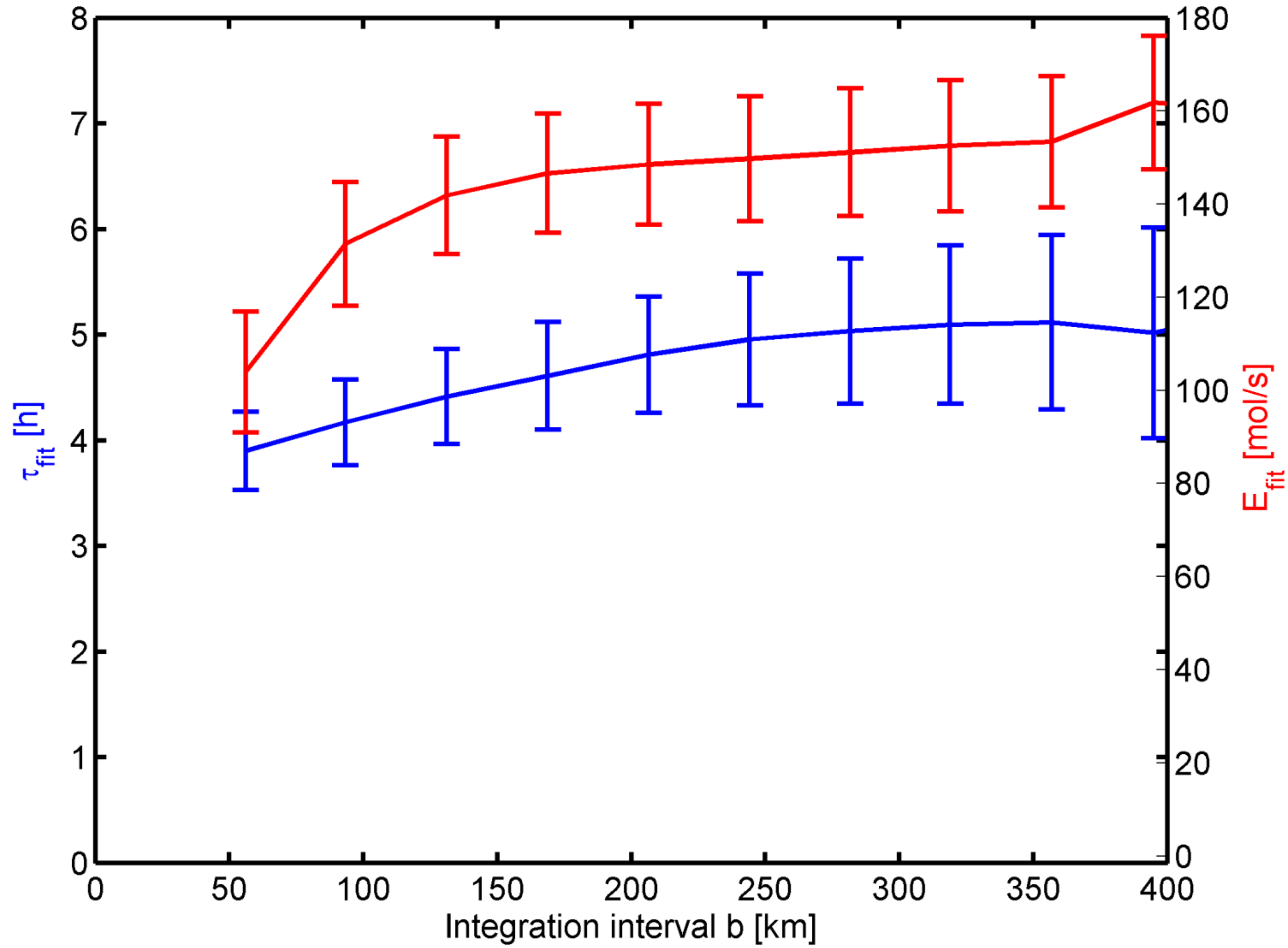
Impact of diurnal cycles. Dependency of fitted lifetime (left) and emissions (right) on the a-priori night-time lifetime and emissions for the synthetic line density model run. A-priori daytime lifetime/emissions was set to 5 hours/1 AU (dotted lines).

Additional slides

Impact of interfering sources on fitted lifetimes (in hours, blue) and emissions (in AU, red), for additional emissions of 10%, 30%, or 50% at 200 km or 100 km distance. For 50% additional emissions at -100 km, the fit performance was deficient.

	-200 km	**+200 km**	**Mean 200 km**	**-100 km**	**+100 km**	**Mean 100 km**
10%	4.0±0.2	5.4±1.0	4.7	3.9±0.5	5.9±0.3	4.9
	1.0±0.0	0.9±0.1	1.0	1.1±0.1	0.9±0.0	1.0
30%	3.0±0.9	7.8±3.6	5.4	2.5±1.6	8.3±1.6	5.4
	1.1±0.3	0.7±0.2	0.9	1.6±0.8	0.8±0.1	1.2
50%	2.3±1.9	13.3±10.7	7.8	-	10.3±3.9	-
	1.3±0.9	0.6±0.3	0.9	-	0.8±0.2	-

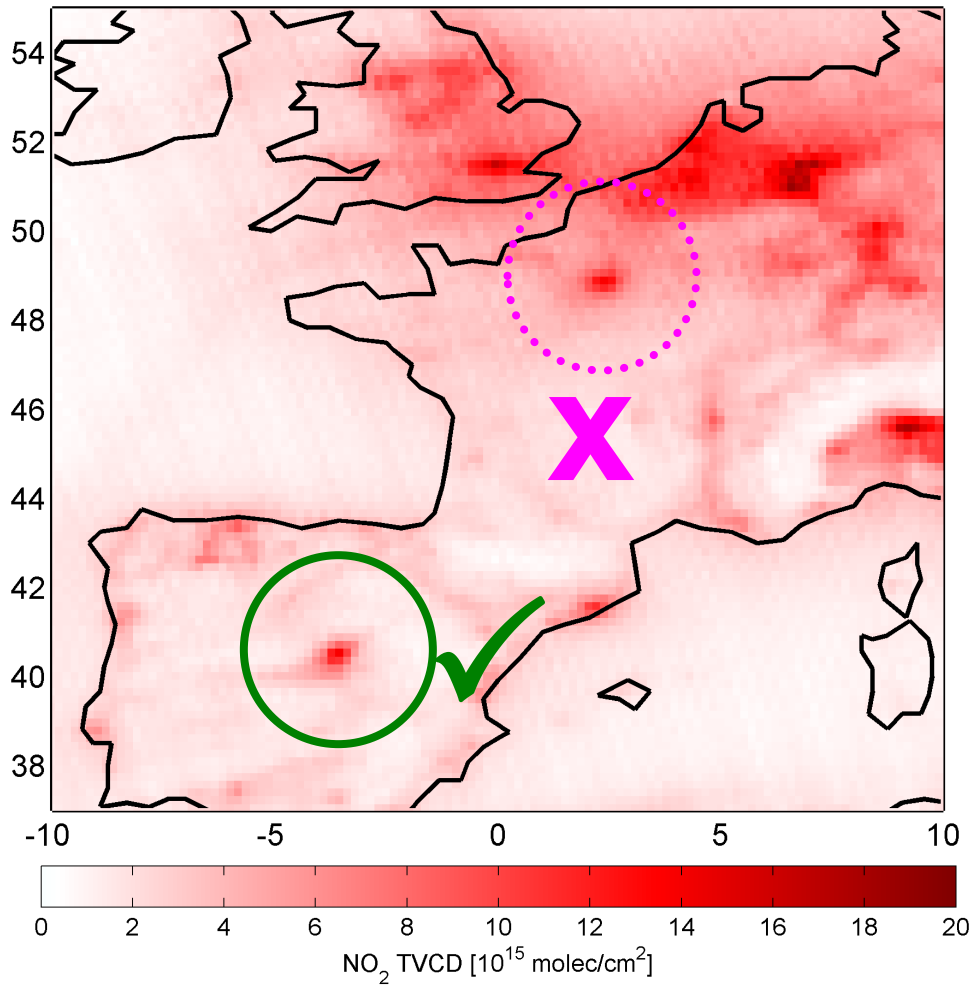
Additional slides



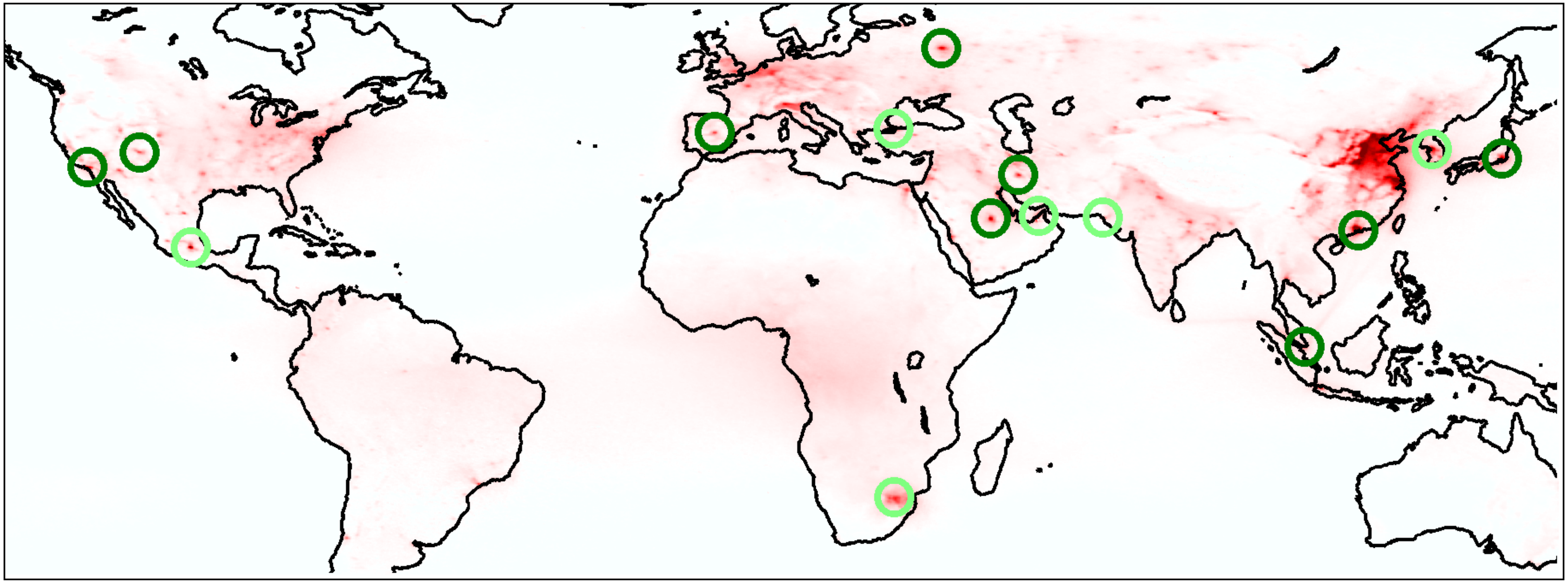
Dependency of fit results for□ t (blue) and *E* (red) on the integration interval *b* for NO2 observations (mean of the fit results from all wind direction sectors) over Riyadh.

Other Point sources

- Definition of point sources via **contrast** and **background homogeneity**:



Other Point sources



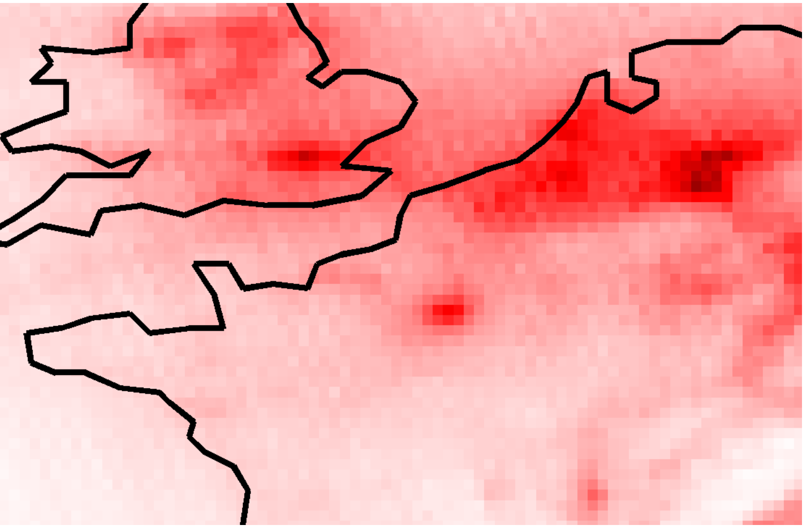
- 15 locations worldwide
- 9 locations with **successful** fit:
  - Mean wind speed > 2 m/s (skips Mexico City)
  - Method works for at least 2 wind direction sectors in each season

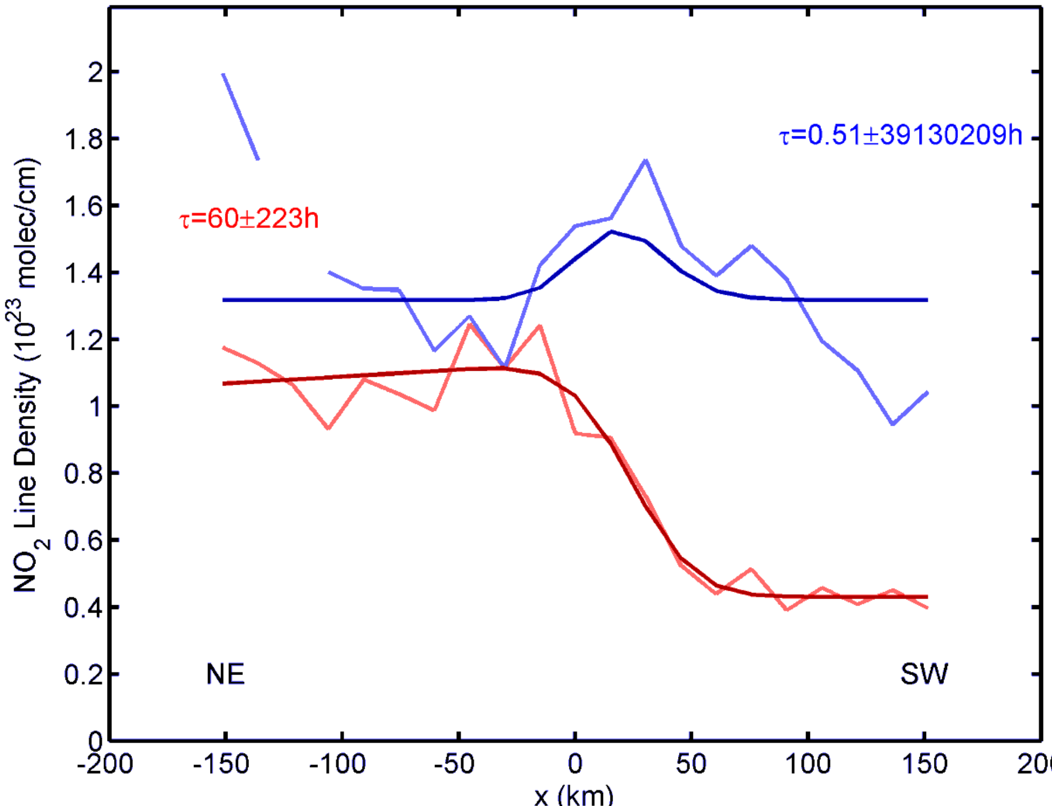
Uncertainties

	τ	E
• NO_2 VCDs		30%
• NO_2/NO_x		10%
• Choice of fit interval	10%	10%
• Choice of wind fields	30%	30%
• Fit confidence interval	10-50%	10-50%
• Fit SME	10-40%	10-40%
• Total (if independent)	~35-60%	~47-63%

- Uncertainties are lowest for Riyadh

MEGAPOLI: What about Paris?





Paris, autumn

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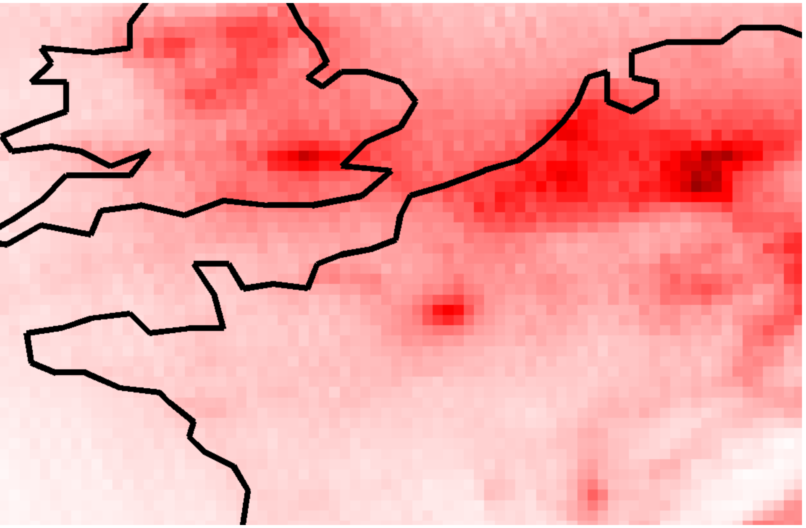
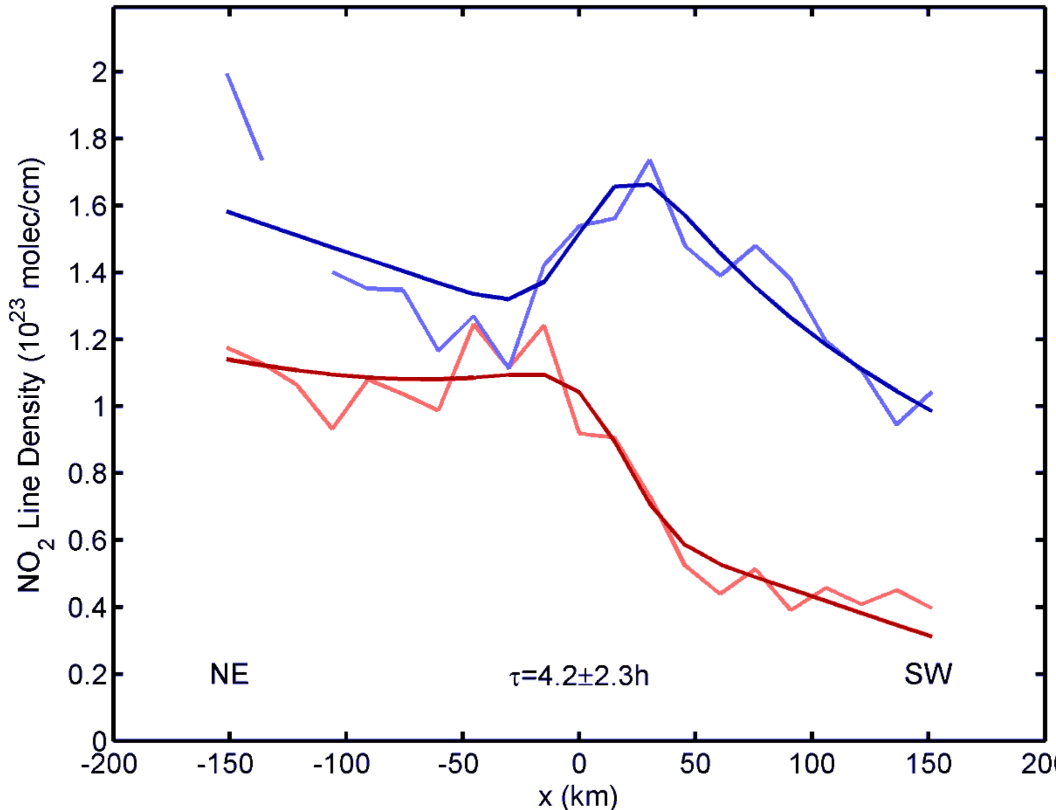


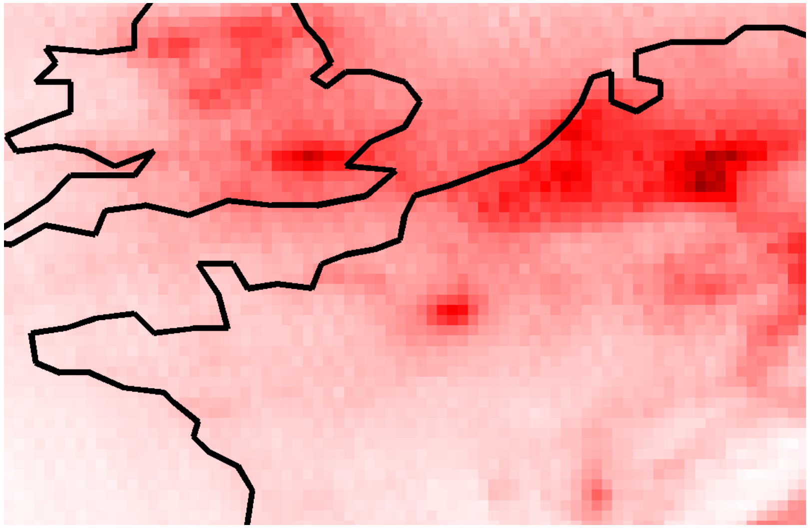


Paris, autumn

Solution:

- Consider **smaller area**
- Allow for (linearly) **varying background**
- Fit **opposite** wind direction sectors **simultaneously**

MEGAPOLI: What about Paris?



Results:

- τ=4.1±1.5 h
- **E=98±47** mol/s

EDGAR: **118** mol/s
(150×150 km^2)

MAXDOAS:
~**53** mol/s (summer)
~**117** mol/s (winter)
(Reza Shaiganfar, personal communication)

MP: ~ **86** mol/s
CITEPA: ~ **104** mol/s
(Hugo van der Gon, personal communication)

Conclusions (I)

- Riyadh (~7M people) is highly polluted!
- High ozone levels
- The large cities in the Middle East should be considered as Megacities, even if <10M!