Trends in tropospheric nitrogen dioxide (NO_2) over megacities in the Mediterranean and Middle East from GOME and SCIAMACHY

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Outline

Introduction

Why study megacities?

- The fraction of world population living in large cities is rapidly increasing (currently, more than 50%).
- Large cities are pollution hot-spots as result of intense traffic, energy use and industrial production.
- Pollution in large cities affects many people.
- Large cities have the potential to export pollution to cleaner regions, in particular if convection is strong.

Why study NO_2?

- High NO_2 concentrations pose significant danger to human health
- NO_2 is one of the few atmospheric polltants under European and international regulation
- NO_2 plays a role in ozone smog formation and acid rain
- Hopefully, it will be possible to estimate NO_x emissions from NO_2 measurements.

Which instruments have been used for this study?

Here, we use **satellite instruments.**

Advantages:

- long and consistent timeseries possible
- global coverage
- makes comparison of different regions possible

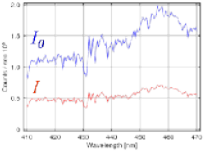
Instruments used for this study:

Instrument	**Equator crossing**	**Global coverage**	**Repeat cycle**	**Ground pixel [km^2]**
GOME	10h30	3 days	35 days	40 $\times$ 320
SCIAMACHY	10h00	6 days	35 days	30 $\times$ 60

The DOAS method

Differencital **O**ptical **A**bsorption **S**pectroscopy – based on *Lambert-Beer*:

$$\underbrace{ln\left(\frac{I_0(\lambda)}{I(\lambda)}\right)}_{spectra} = \underbrace{\int_S \sum_i \rho_i(s)\,\sigma_i'(\lambda, s)\,ds}_{absorption\ term} + \underbrace{\sum_k a_k \lambda^k}_{polynomial} + \underbrace{r(\lambda)}_{residual}$$



– trace gases
– ring effect

– broad-band effects

- $\sigma_i' + \sigma_i^b = \sigma_i$: Differential + broad-band = total absorption
- Main inputs:
  - I, I_0, the actual and background spectra from the instrument
  - σ_i', the reference absorption cross sections, measured in lab
- Main result: $SC_i = \int_S \rho_i(s)ds$, the *slant column* of each trace gas i

Accounting for stratospheric NO_2

- The satellite instruments measure the **total slant column density** SCD_{tot} of a trace gas, which is the integral along the average light path (in molecules per cm^2)
- This signal consists of a tropospheric and a stratospheric contribution

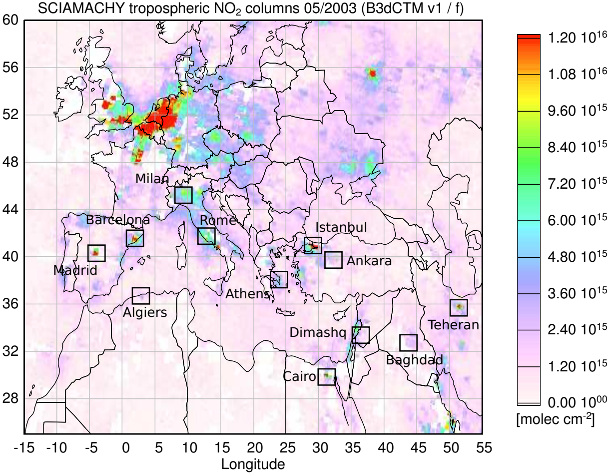
$$SCD_{tot} = SCD_{trop} + SCD_{strat}$$

- Here, we calculated SCD_{strat} from model data (B3dCTM):
  - Global model values were scaled to the level of the satellite measurements in a *reference sector* over the Pacific Ocean
  - Application of an *air mass factor* to construct SCD_{strat}
  - Subtract this value from SCD_{tot} to yield SCD_{trop}

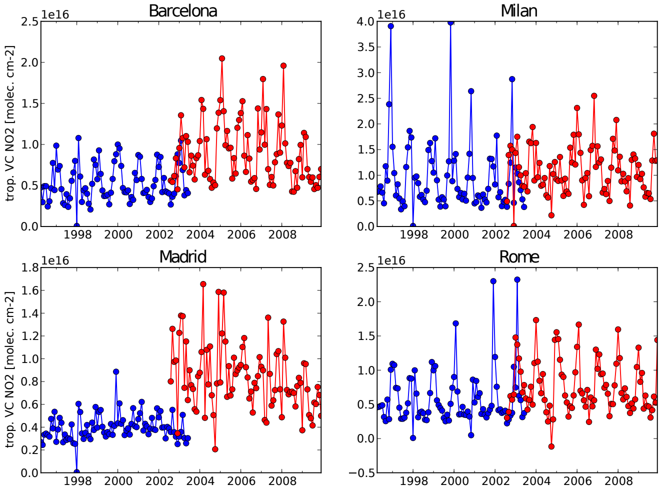
The combined GOME and SCIAMACHY timeseries

Overview

- Grid sat. pixels on .125° × .125°
- Calculate monthly averages
- Define city regions (not shown to scale here!)
- For each month, calculate the average of each city region individually



Combined timeseries from GOME and SCIAMACHY



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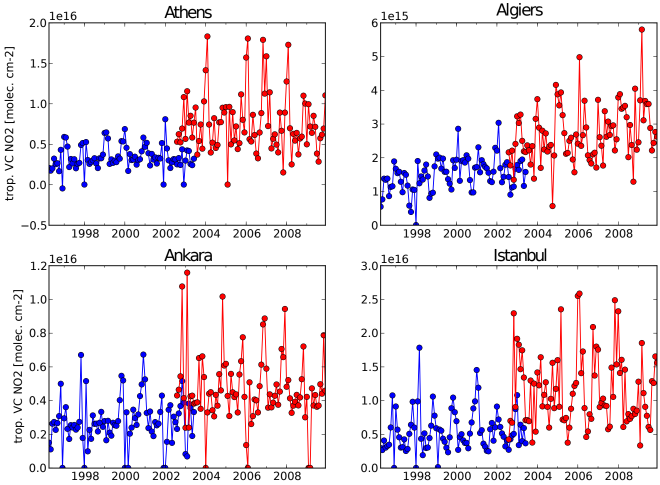

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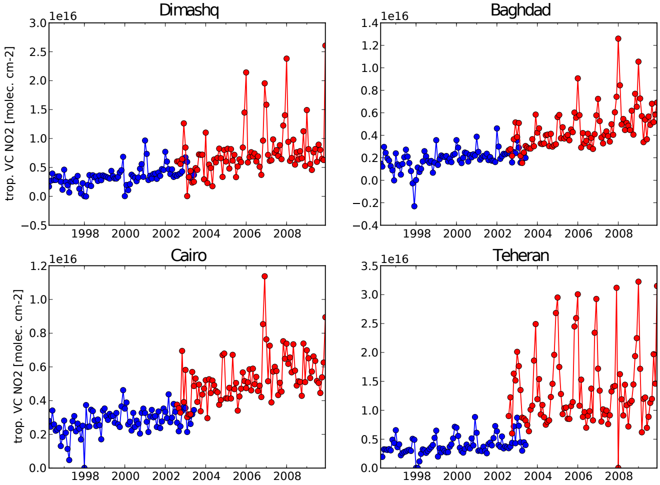


Summary

- Most investigated cities show upward tendencies
- Trends for eastern and North African megacities seem to be a lot stronger than those for western cities
- Most cities show a huge gap between the GOME and SCIAMACHY timeseries
- **Quantitative estimation of trends not feasible!**

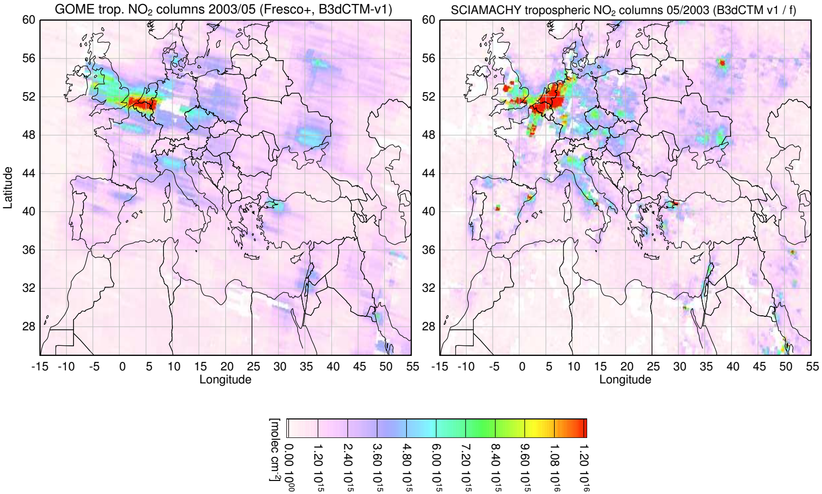
Combining GOME and SCIAMACHY:

’Deconvolution’ of GOME data

The problem of differing pixel sizes

- GOME and SCIAMACHY have very different pixel sizes
- For averages over large areas, this is no big problem (see *Richter et al., 2005*)
- For point sources (like cities), this leads to a relatively diluted signal in the GOME data: The same total amount of NO_2 is averaged over a larger area.
- This leads to inconsistencies in timeseries spanning both GOME and SCIAMACHY.

The problem of differing pixel sizes



What has been done so far?

Most previous studies artificially reduced the spatial resolution of SCIAMACHY measurements:

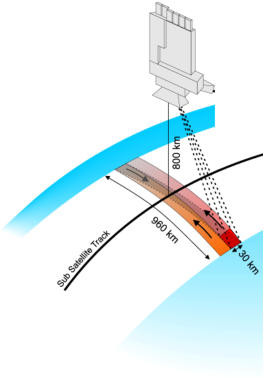
Publication	Time period covered
van der A et al., 2008	1996-2006
Konovalov et al., 2010	1996-2008

City	Annual trend *van der A.*	Annual trend *Konovalov*
Baghdad	—	1.7 $\pm$ 0.7 %
Barcelona	—	3.7 $\pm$ 0.8 %
Cairo	1.3 $\pm$ 1 %	—
Tehran	6.5 $\pm$ 1 %	4.0 $\pm$ 0.8 %

Another approach was used by *Konovalov et al., 2006*:

- SCIAMACHY measurements were artificially convoluted with a Gaussian function to yield deconvolution factors for GOME measurements.

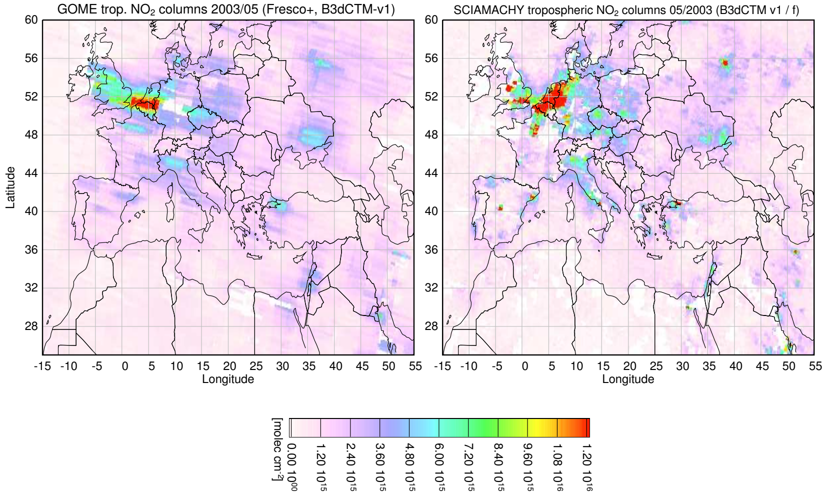
Making use of SCIAMACHY backscan measurements



Impose the spatial structure of SCIAMACHY measurements on GOME data:

- daily global maps of SCIAMACHY back and forward scan measurements
- daily global maps of $SCIA_{forw}$ / $SCIA_{back}$
- monthly means of these maps
- monthly climatology for the years 2004-2009 from these maps
- monthly global maps of GOME measurements
- Multiply each such map with the corresponding map from the correction factor climatology

The impact of the SCIAMACHY back scan method



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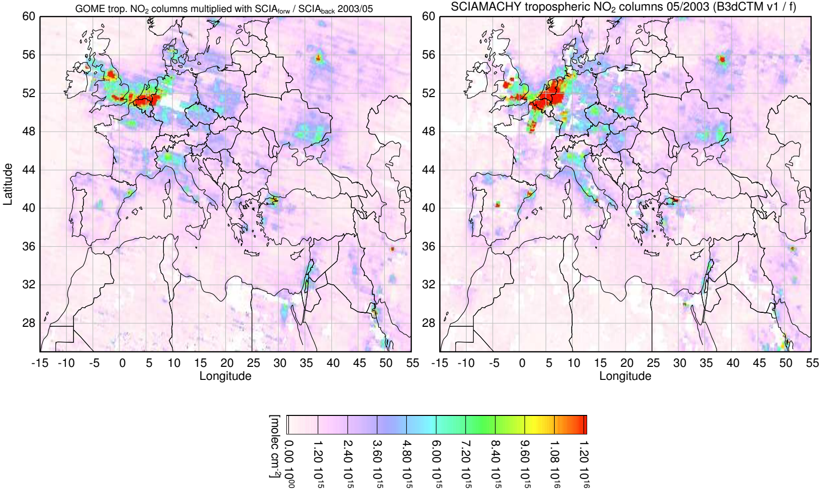
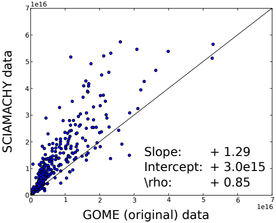
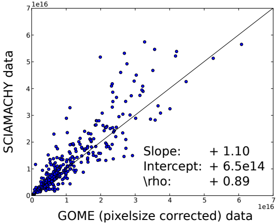


Comparison corrected / original dataset

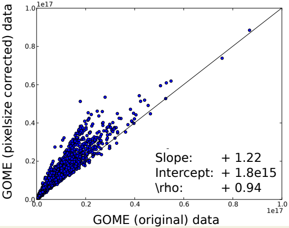
GOME (original) vs. SCIAMACHY
Aug. 2002 - Jun. 2003



GOME (pixel-size-corrected) vs. SCIAMACHY
Aug. 2002 - Jun. 2003



GOME (original) vs. GOME (pixel-size-corrected)
Apr. 1996 - Jun. 2003



GOME (original) vs. GOME (pixel-size-corrected)
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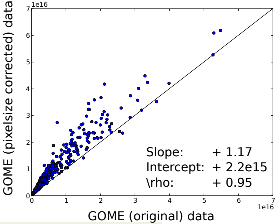
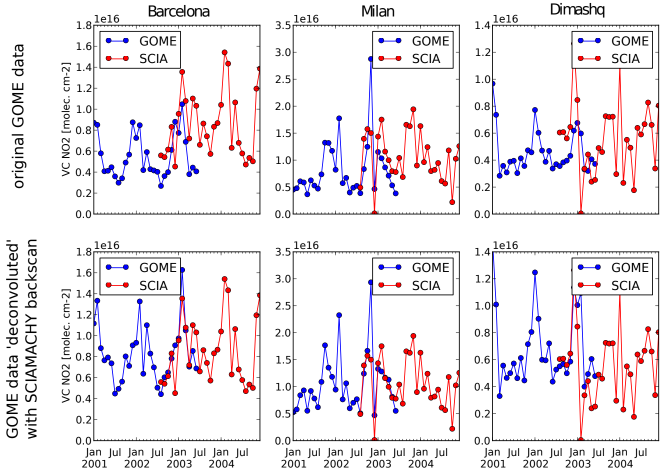
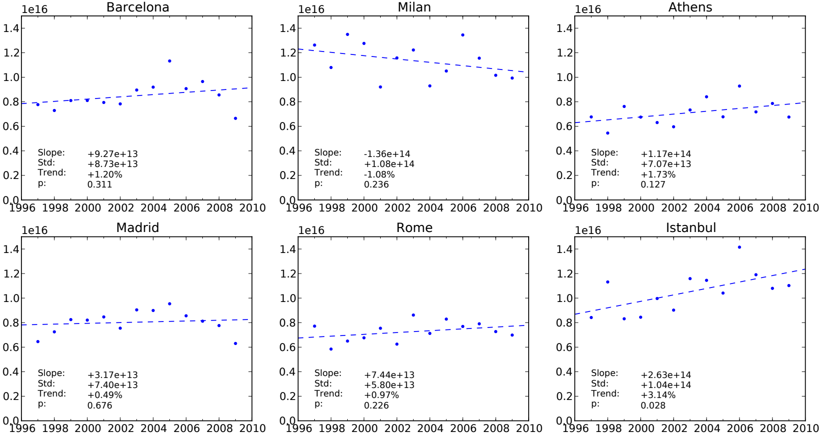


The common period of GOME and SCIAMACHY: 2002/08 – 2003/06



Linear trends over selected megacities



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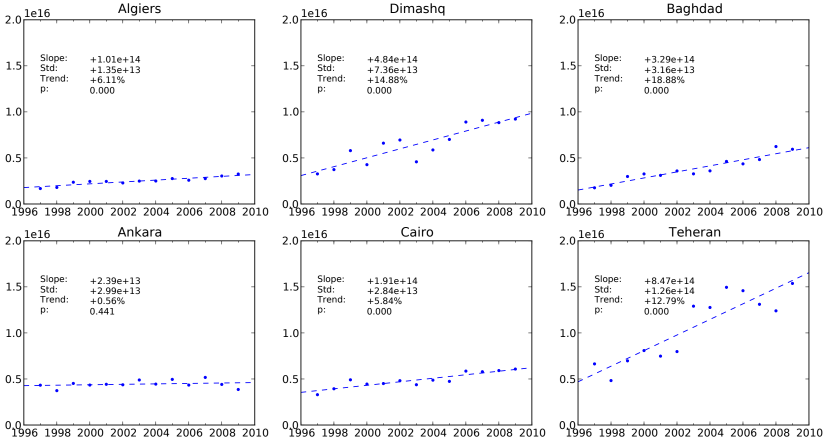

Comparison to previous studies

Annual growth rate (in %) over selected cities:

City	This study	van der A.	Konovalov
Baghdad	18.9 ± 1.8 %	—	1.7 ± 0.7 %
Barcelona	*1.2 ± 1.1 %*	—	3.7 ± 0.8 %
Cairo	5.8 ± 0.9 %	1.3 ± 1 %	—
Tehran	12.8 ± 1.9 %	6.5 ± 1 %	4.0 ± 0.8 %

- Observed trends are significantly higher than in previous studies
- This is because previously,
  - the poor spatial resolution of GOME measurements has been used as a reference
  - the investigated regions were larger

Difference in overpass time between GOME and SCIAMACHY

Additional problem: **difference in overpass time** between GOME and SCIAMACHY.

At $+45^\circ$ latitude:

GOME at 11^{05}h

SCIAMACHY at 10^{36}h

- NO_2 changes significantly during that time!

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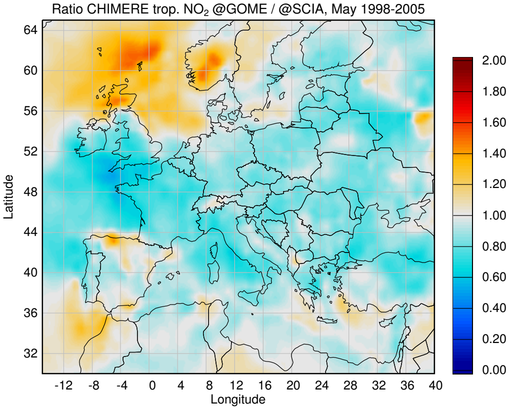
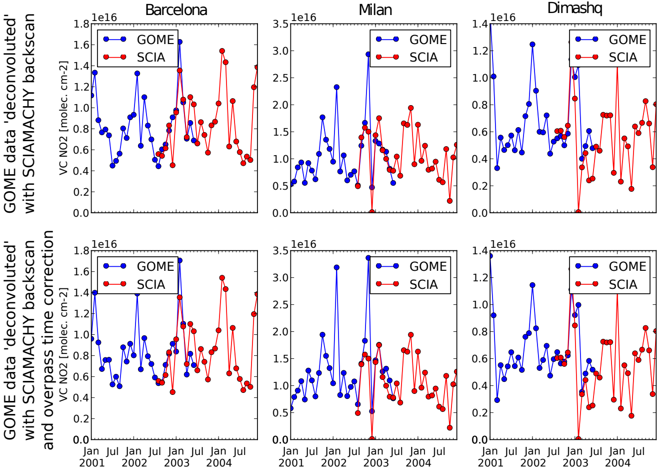


Correcting for the difference in overpass time between GOME and SCIAMACHY



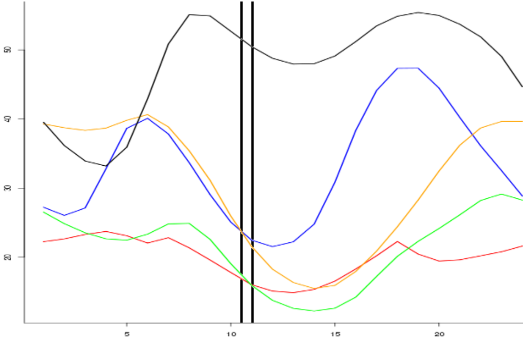
Which model to trust?

Correcting for the diurnal cycle using model data looks promising . . .

But: **Which model to trust?**

Diurnal cycle of surface NO_2 concentrations over BeNeLux pollution hotspots:

- in-situ measurements (black)
- EMEP (red)
- Eurad (blue)
- Bolchem (orange)
- Chimere (Green)



Summary

- The different pixel sizes of GOME and SCIAMACHY pose big problems when investigating point sources
- This problem was attacked by imposing the spatial structure of SCIAMACHY data on GOME measurements by making use of SCIAMACHY backscan measurements
- While the approach makes things better, it far but solves the problem.

- An additional problem is introduced by the difference in overpass time between GOME and SCIAMACHY, and the unknown changes in tropospheric NO_2 during that time.
- These changes can be investigated using model data
- Different models show different diurnal cycles
- Which model to trust?

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- The different pixel sizes of GOME and SCIAMACHY pose big problems when investigating point sources
- This problem was attacked by imposing the spatial structure of SCIAMACHY data on GOME measurements by making use of SCIAMACHY backscan measurements
- While the approach makes things better, it far but solves the problem.

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- These changes can be investigated using model data
- Different models show different diurnal cycles
- Which model to trust?

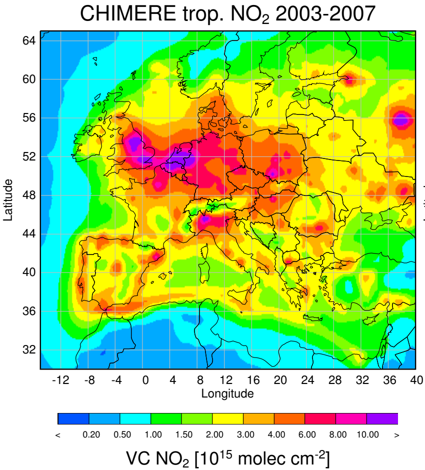
Comparison to model data:

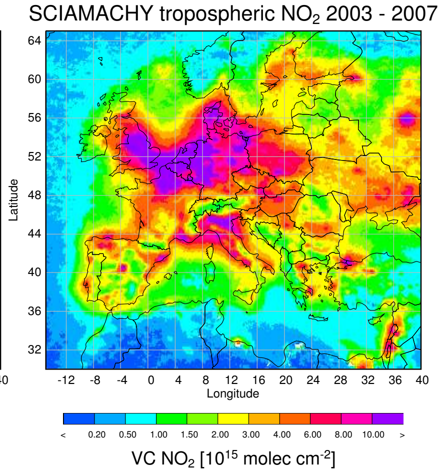
CHIMERE

Basics about the CHIMERE data used in this study

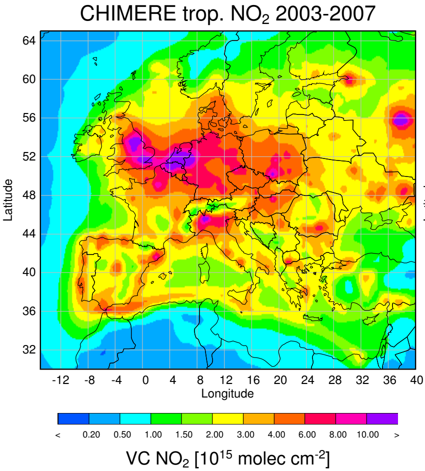
- Regional model developed at *LMD*
- More information at
  `http://www.lmd.polytechnique.fr/chimere/`
- This model run
  - covers the period 1998 – 2007
  - has a timestep of one hour
  - runs on a 0.5° × 0.5° grid
  - is driven by EMEP 0.5° × 0.5° emissions data
  - only covers the lowest ˜ 5km of the troposphere

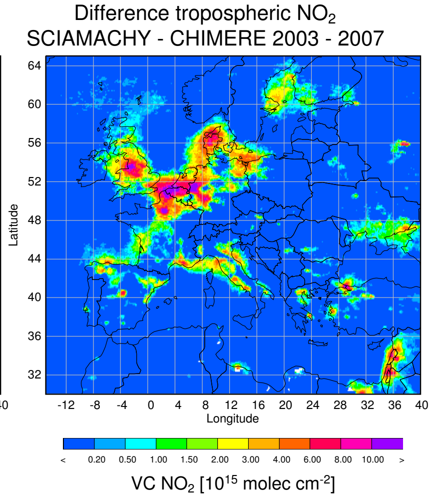
Tropospheric NO_2 from CHIMERE (2003 – 2007)



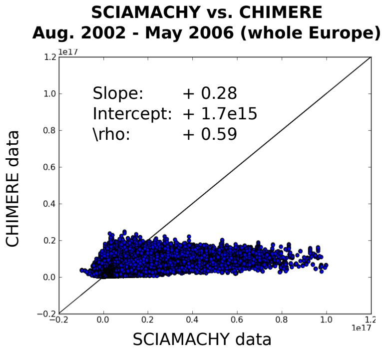


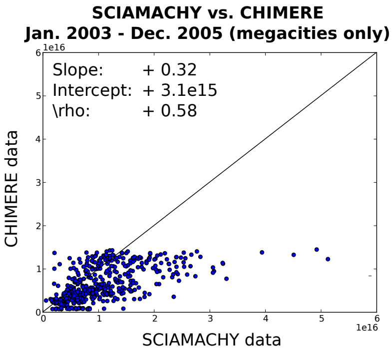
Tropospheric NO_2 from CHIMERE (2003 – 2007)



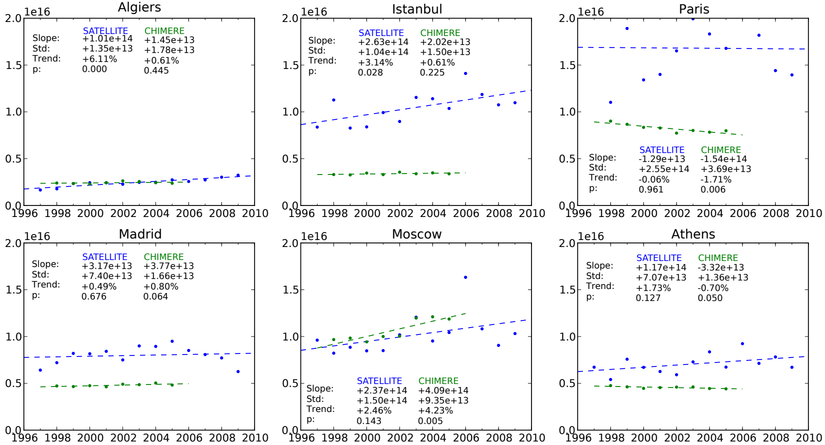


Correlation between SCIAMACHY and CHIMERE





Annual trends



Summary

- CHIMERE values over megacities are far too low
- This is effect of the coarse model resolution
- Low values are in accordance to previous studies
- Correlation with SCIAMACHY measurements is disappointing
- Where significant, trends from CHIMERE are stronger than from satellite

Future work

Future work

- Calculate trends for tropospheric NO_2 from GOME2 and OMI and compare these to the trends from SCIAMACHY
- Study the influence of city boundary definition on the derived trends
- Study the influence of different cloud selection criteria on the derived trends
- Look for nonlinear trends
- Look at the GOME deconvolution issue in more detail:
  - Study effect of different Gauss kernels on the derived trends
  - Correct for the GOME *narrow swath mode*
- Use vertical NO_2 profiles from Chimere to create a dataset of daily air mass factors

Acknowledgements

- BMS for providing B3dCTM data
- Augustin Collette (INERIS) for providing CHIMERE data
- Igor Konovalov for sharing his insight on trend analysis
- TEMIS for providing FRESCO+ data
- ESSRES for funding my PhD work
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