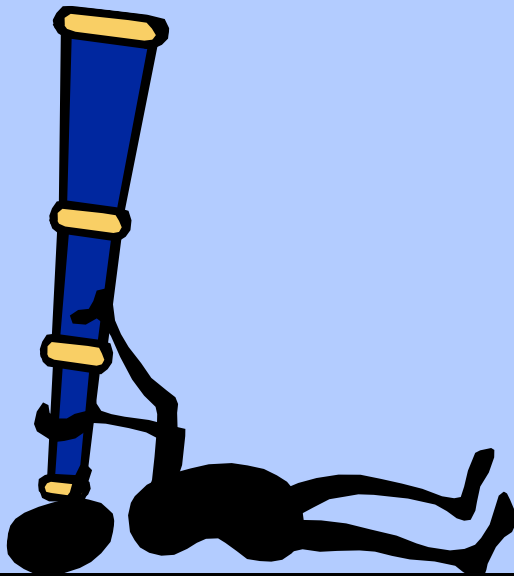


Ground Based Observations of Tropospheric NO₂

*Algorithm development
and validation*



Tim Vlemmix | Ankie PETERS

The need for ground based column observations

The MAX-DOAS technique

Development of a retrieval model

Sensitivities and error analysis

Comparisons

Summary and Conclusions

Overview

The need for ground based column observations

The MAX-DOAS technique

Development of a retrieval model

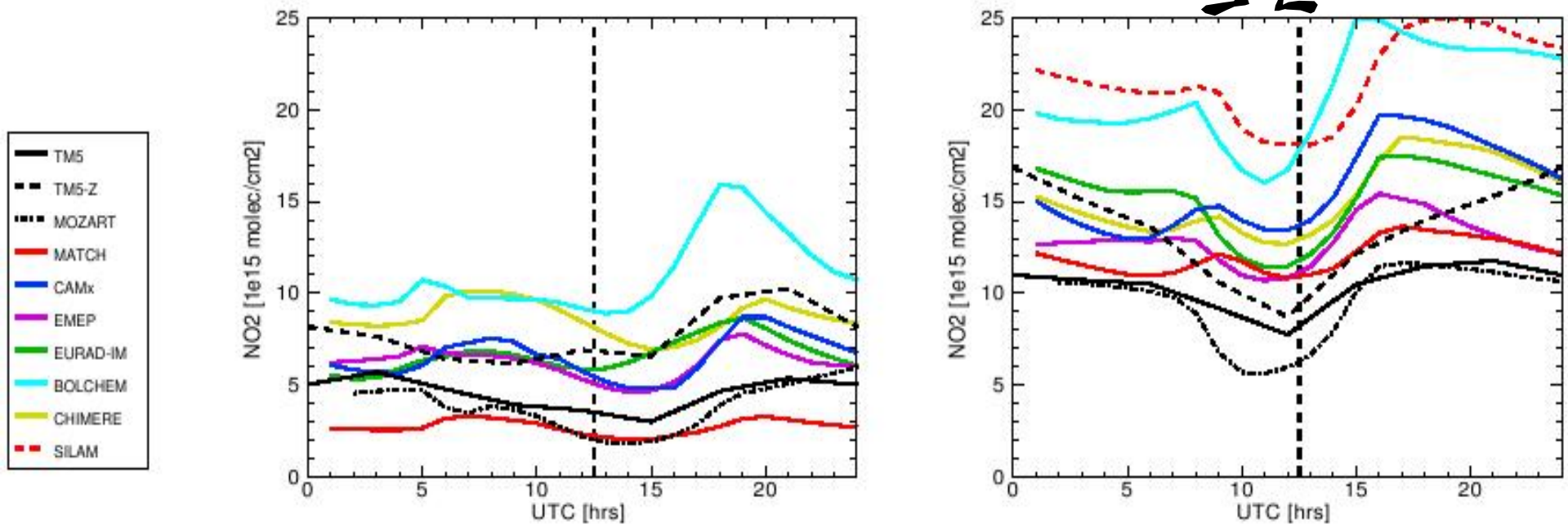
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Air Quality Model Validation

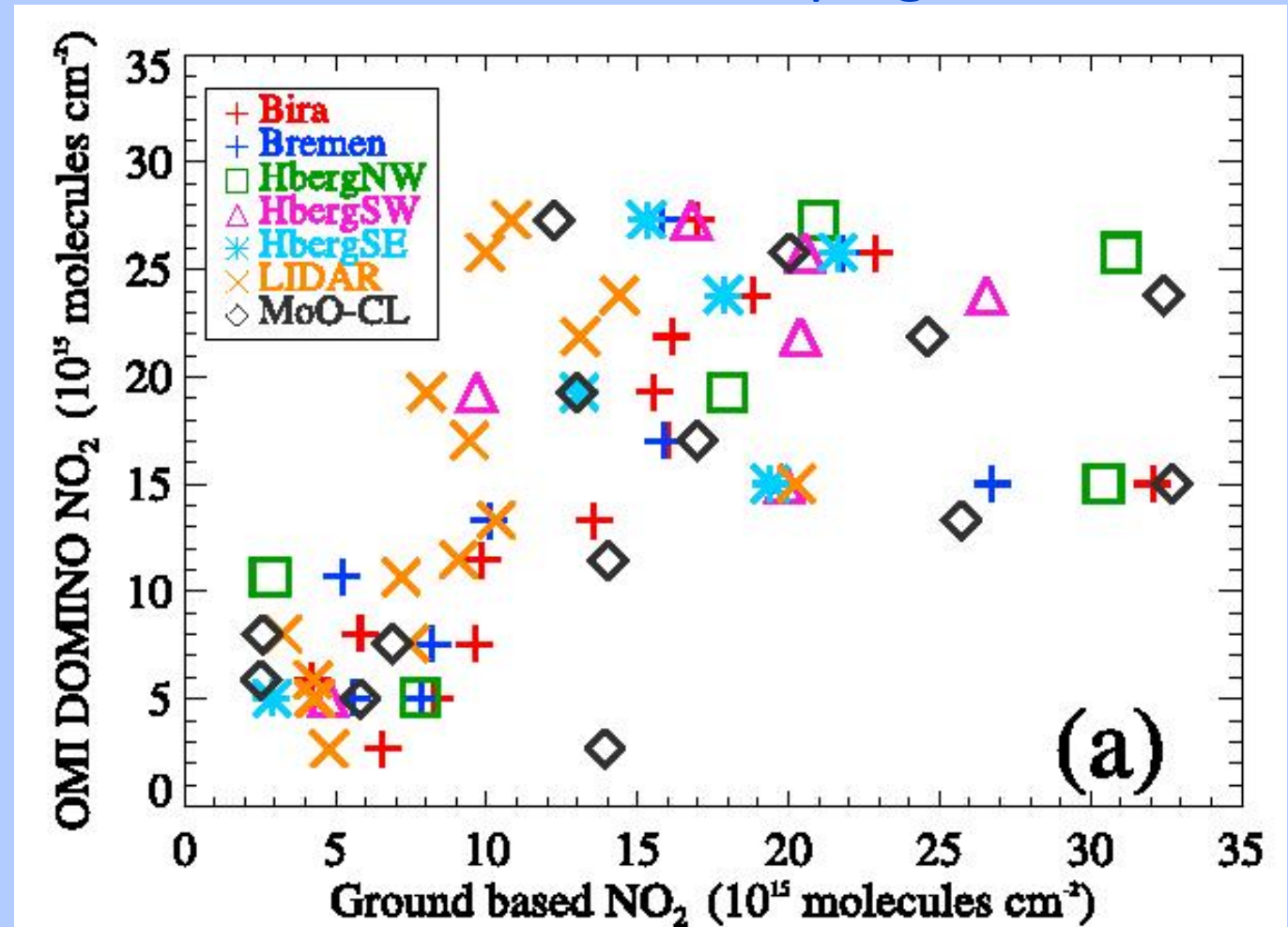
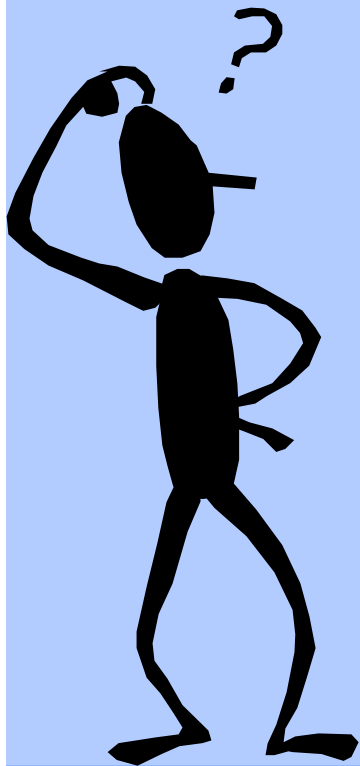


Monthly averaged diurnal cycle Aug. & Dec.

Huijnen et al., 2010

Large differences between Air Quality Models

Dandelions Campaign 2006

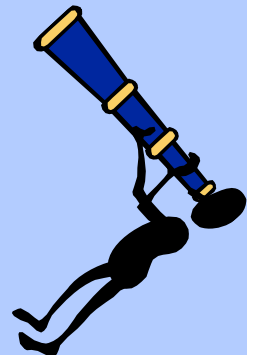
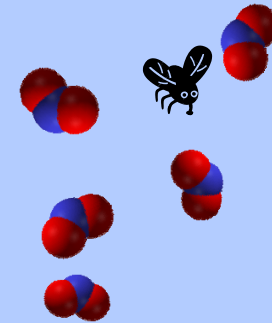
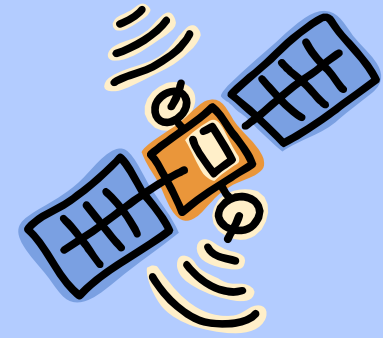


Hains et al., 2009

Satellite validation results pose many questions

Issues Satellite Validation of NO₂

- MAX-DOAS at Dandelions: geometrical inversion, no radiative transfer modeling. Crude assumptions
- MAX-DOAS is more sensitive towards surface
- Satellite is more sensitive away from surface
- Validation critically depends on knowledge of profile shape, no information from satellite
- Spatial inhomogeneity complicates comparison





Million Dollar Question....



.... can we retrieve the NO_2
profile with a MAX-DOAS???



The need for ground based column observations

The MAX-DOAS technique

Development of a retrieval model

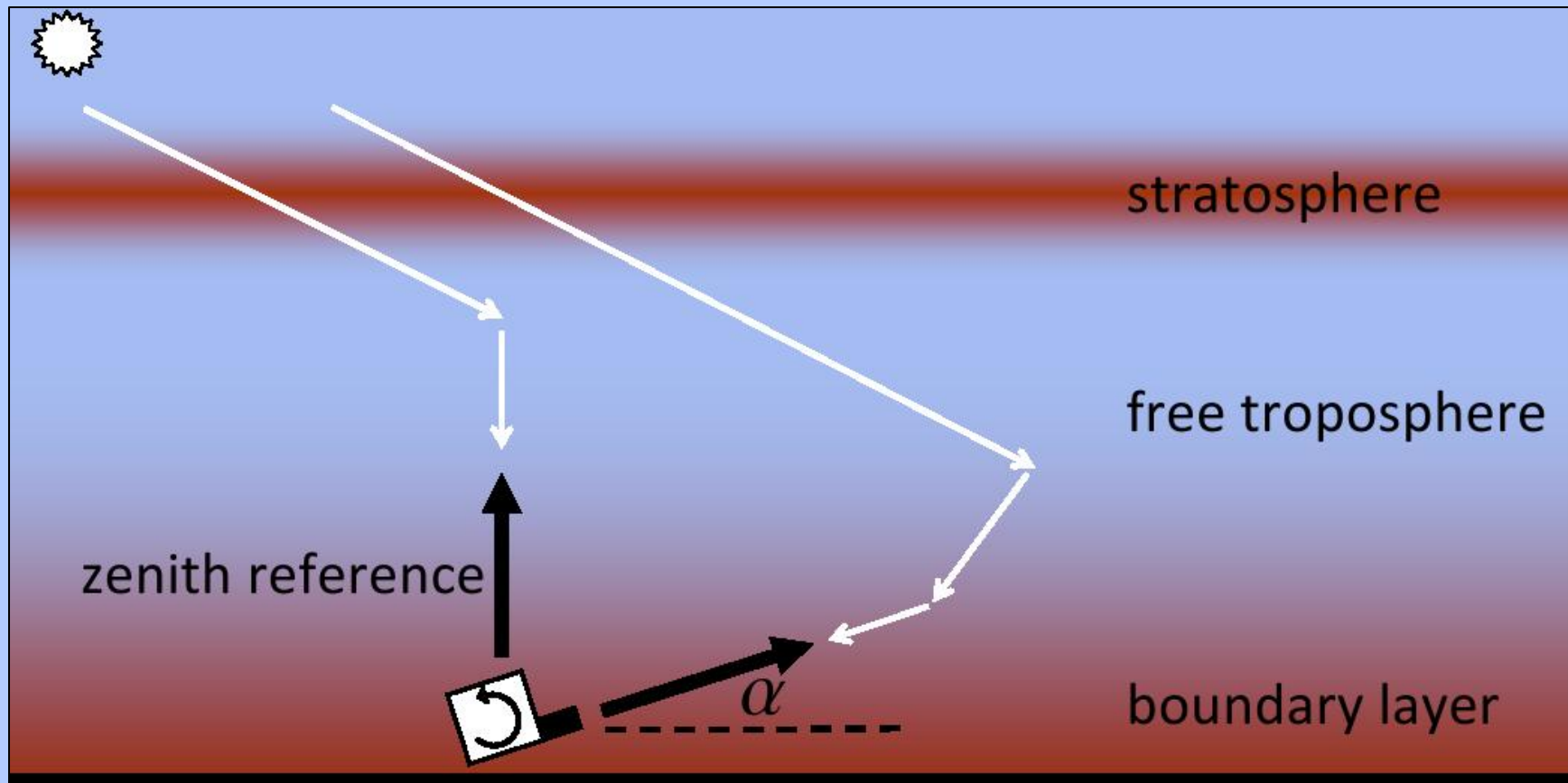
Sensitivities and error analysis

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>> **Differential Slant Columns** <<

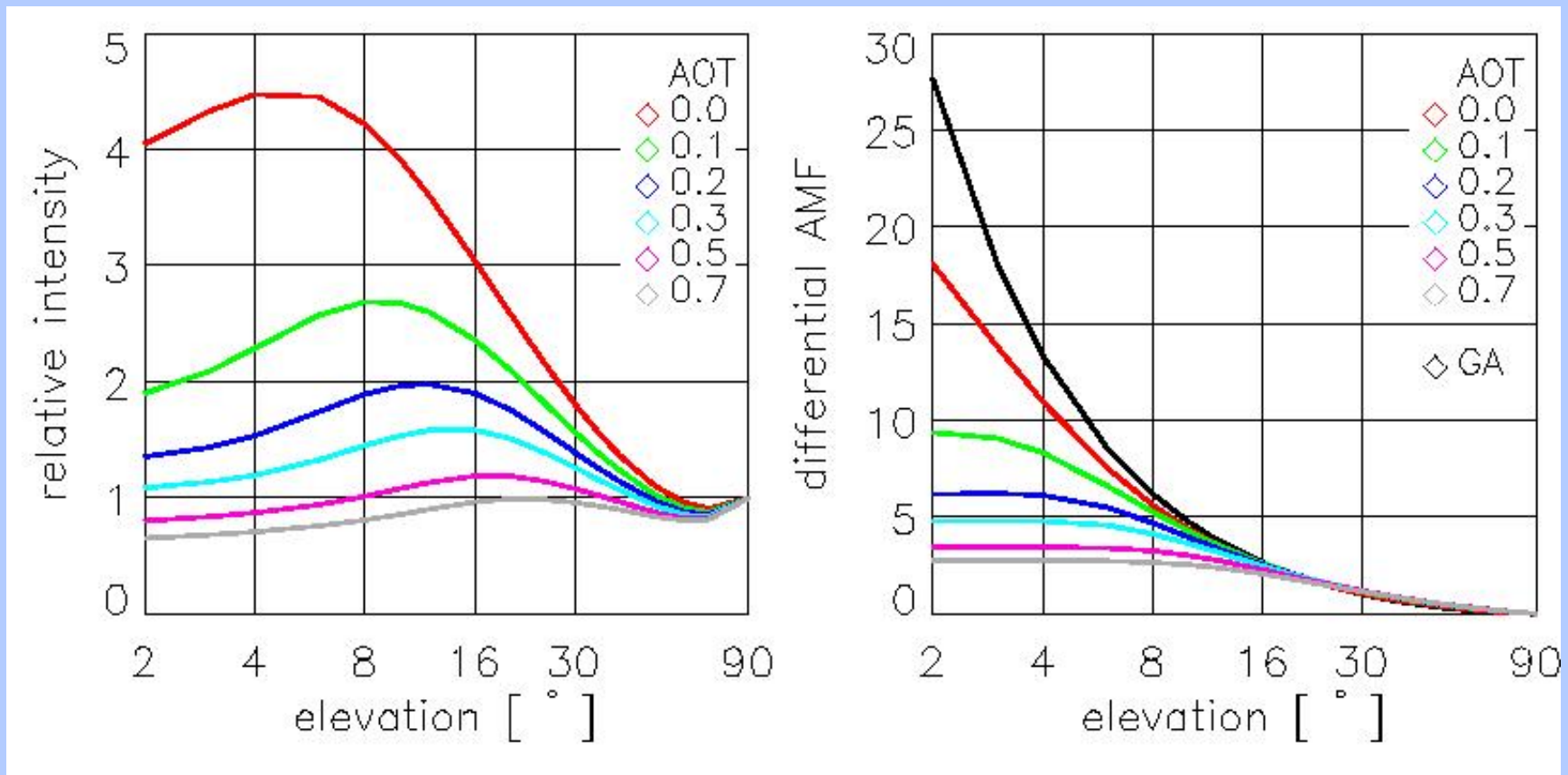
The MAX-DOAS Technique

MAX-DOAS Measurement Quantities:

- Differential Slant Columns of NO₂
- Differential Slant Columns of O₄ >> light path
- Relative Intensities (or relative radiances)

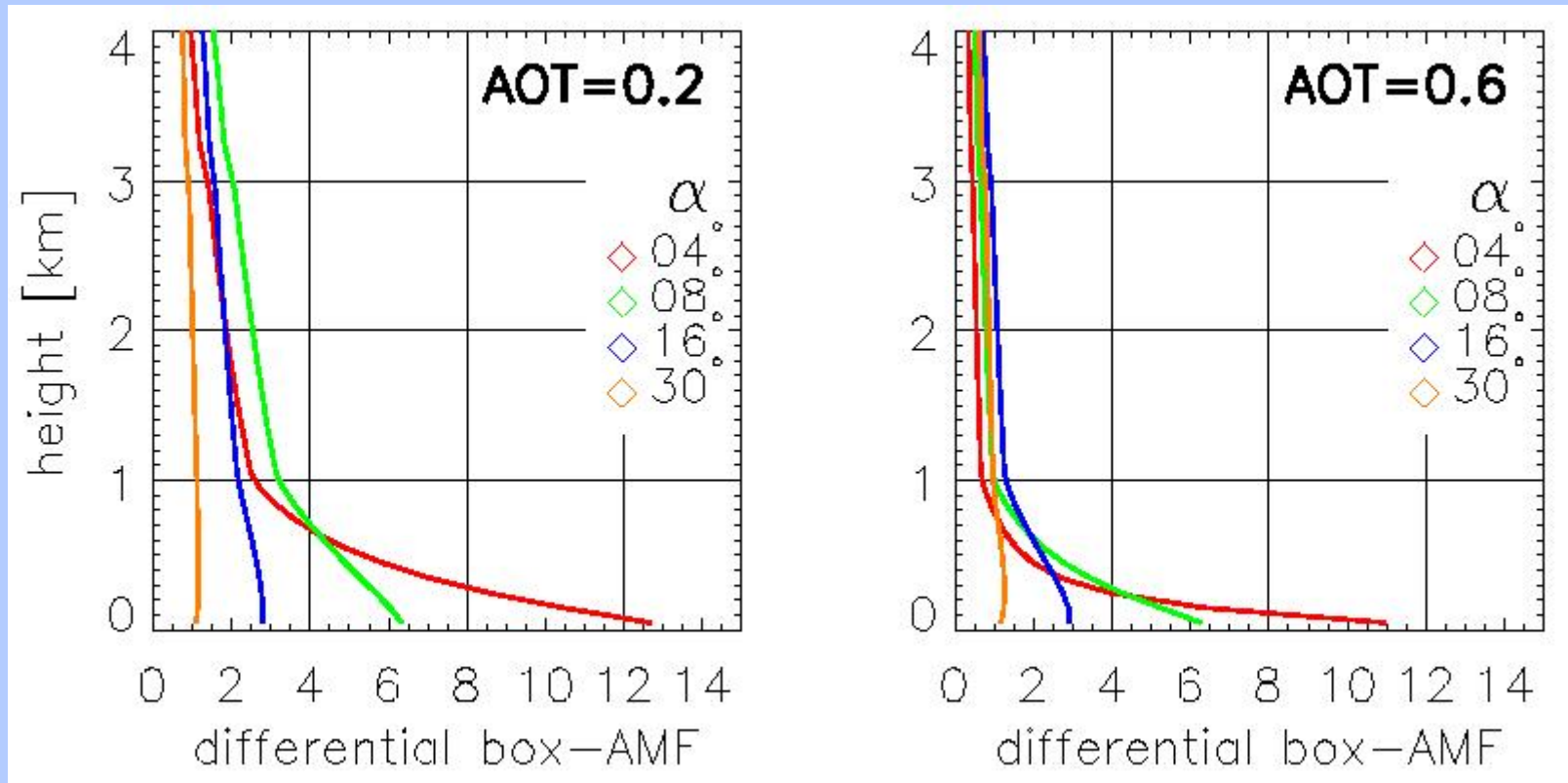
$$\Delta N_{\alpha}^S = N_{\alpha}^S - N_{90^{\circ}}^S$$

$$I_{\alpha}^{rel} = \frac{I_{\alpha}}{I_{90^{\circ}}}$$

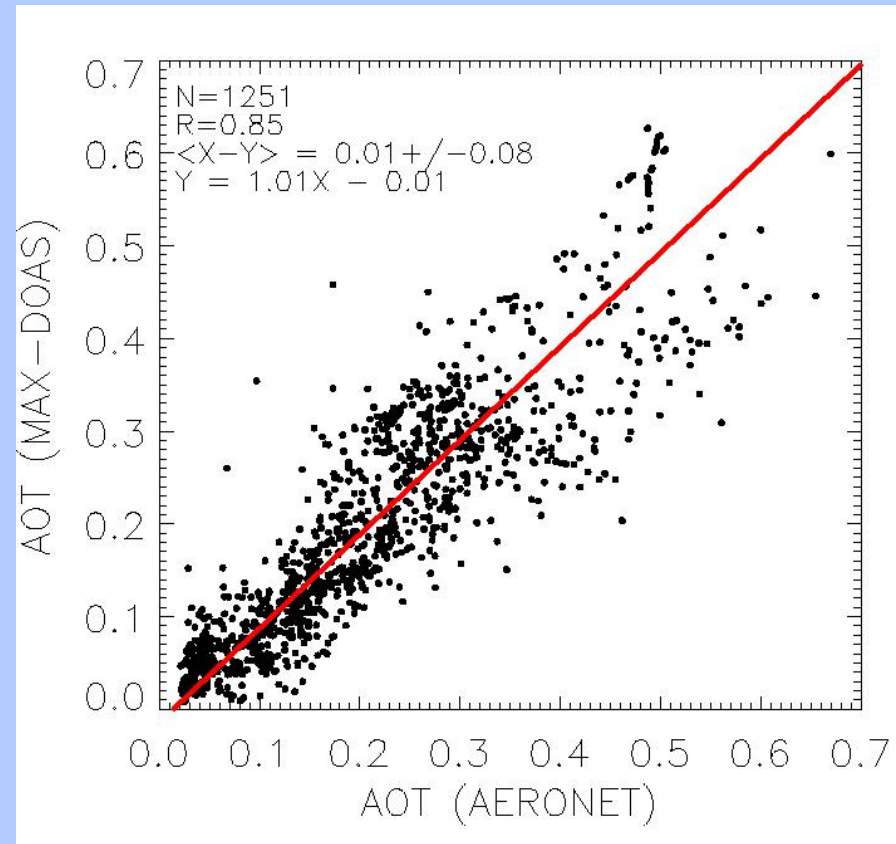
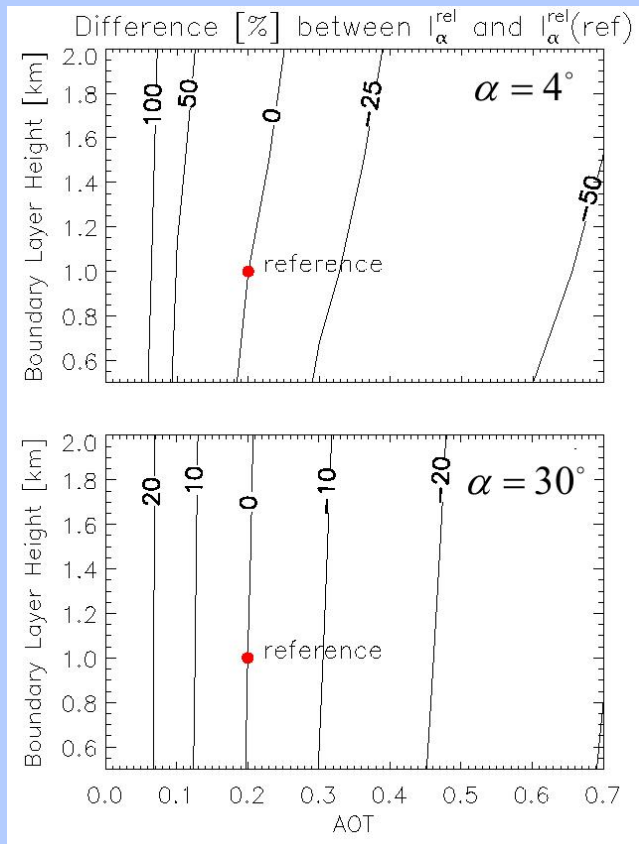


Large Impact Aerosols on AMF >> include in retrieval

Vertical sensitivity to NO_2 depends on elevation

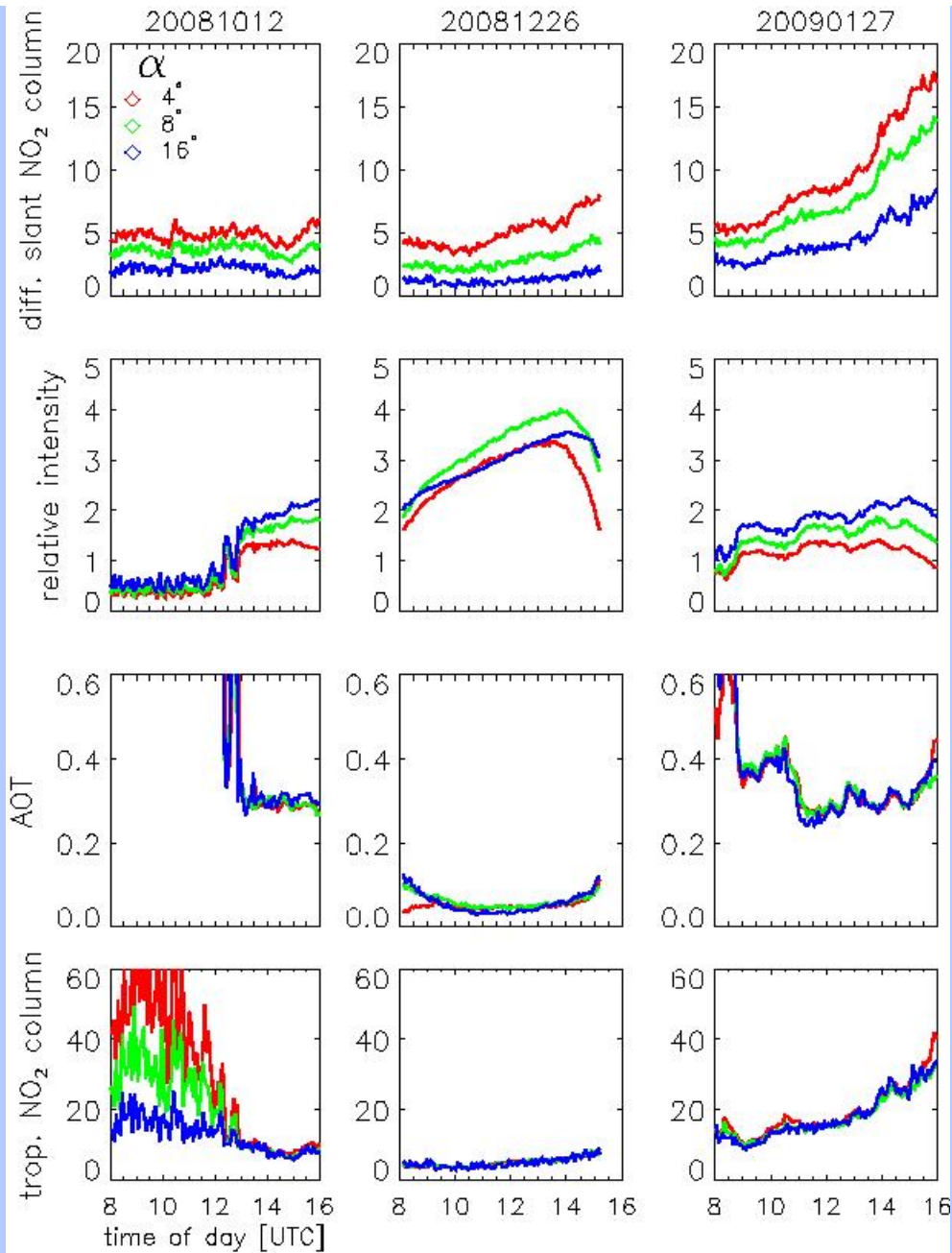


Potential and Limitations of MAX-DOAS Profiling



- Relative Intensities are quite insensitive to aerosol profile
- O_4 is more sensitive to profile shape in lowest 1-2 km, but almost insensitive above

Relative Intensity and O_4 are complementary



Largest remaining
(controllable) error sources:

- >> NO₂ profile
- >> Aerosol profile

Tropospheric NO₂ column retrieval with aerosol correction

The need for ground based column observations

The MAX-DOAS technique

Development of a retrieval model

Sensitivities and error analysis

Comparisons

Summary and Conclusions

Overview

Algorithm Development

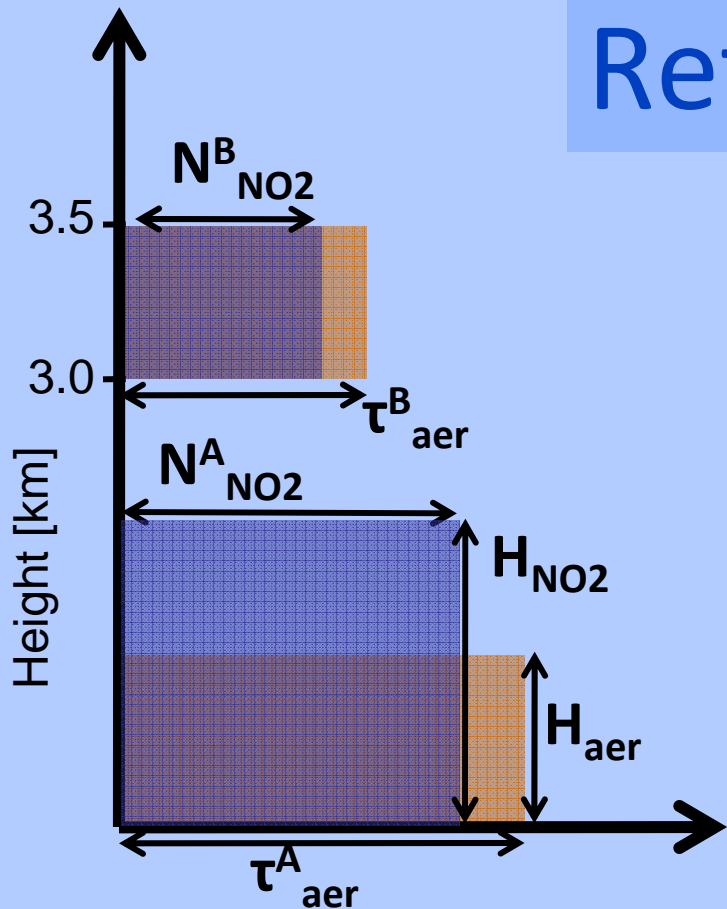
Aim: Exploit the potential of MAX-DOAS to retrieve the *vertical distribution* of aerosols and NO_2

Questions:

- *Which is the optimal set of model parameters?*
- *Is it possible to separate boundary layer NO_2 from free tropospheric NO_2 ?*
- *Which measurements are most essential?*

Column retrieval is not independent of profile

Retrieval Model



Model Assumptions: → Sensitivity study

- Homogenous mixing of aerosols and NO₂ in surface layers with variable height
- Fixed height of “tropospheric” layer [careful interpretation retrieval outcome!]

Options:

- 4P or 6P
- Incl. / Excl. Relative Intensity or O₄
- UV/VIS

Cost Function:

$$C = \sum_{i=1}^4 \left[\frac{C_T \cdot \Delta N_{\alpha_i}^{NO_2} - \Delta N_{\alpha_i}^{NO_2}}{\sigma_{\alpha_i}^{NO_2}} \right]^2 + \left[\frac{C_T \cdot S \cdot \Delta N_{\alpha_i}^{O_4} - \Delta N_{\alpha_i}^{O_4}}{\sigma_{\alpha_i}^{O_4}} \right]^2 + \left[\frac{I_{\alpha_i}^{rel} - I_{\alpha_i}^{rel}}{\sigma_{\alpha_i}^{rel}} \right]^2$$

Finding a solution:

- Calculate CF for whole domain of the 6 parameters in the LUT
- Zoom-in where $CF < 1.5 \times \min(CF)$

Solution consists of:

- 6 parameters
- error estimate for each of 6 parameters
- separate error estimate for “derived” parameters: total NO_2 , total AOT, concentrations

Issues in finding the solution:

- How to organize the zooming in process?
- Resolution and computing time are strongly related (linear interpolation in 6 dimensions)
- Is it possible to uncouple the aerosol and NO₂ retrieval?
 - >> NO₂ does almost not affect the aerosol retrieval, except for some extreme cases, but (uncertainties in the) aerosols do affect the (uncertainties in the) NO₂ retrieval <<

The need for ground based column observations

The MAX-DOAS technique

Development of a retrieval model

Sensitivities and error analysis

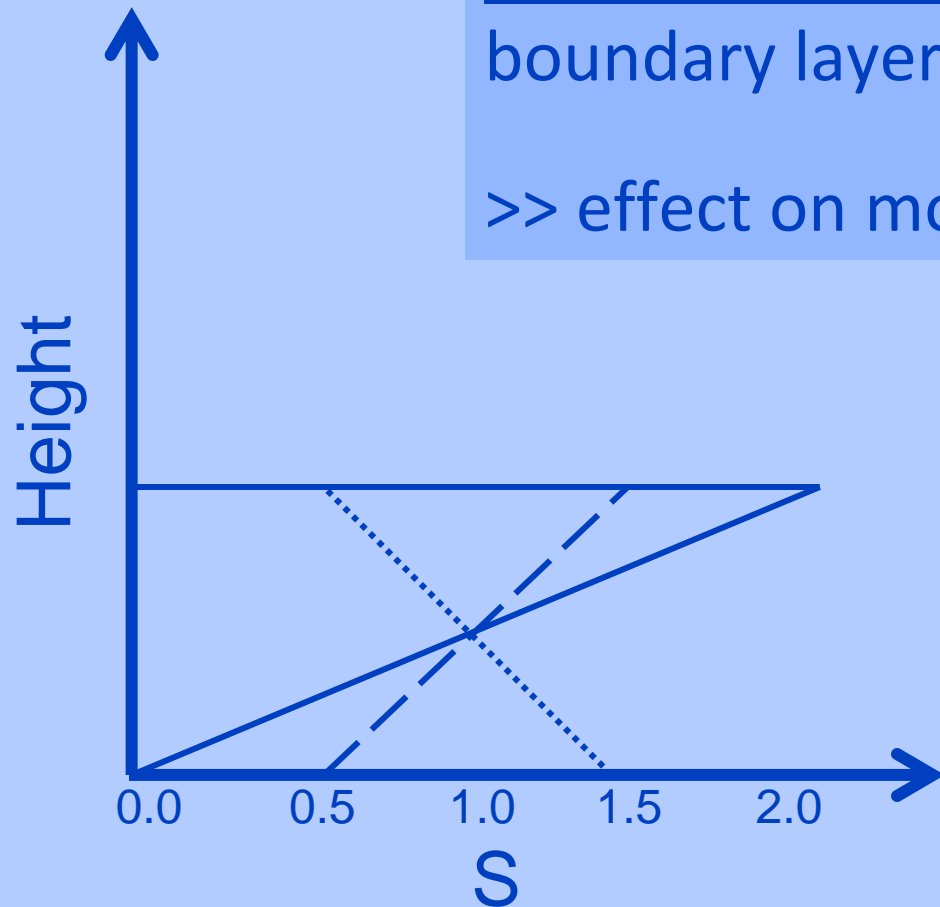
Comparisons

Summary and Conclusions

Overview

Sensitivity study #1 : non-uniform boundary layer for aerosols or NO_2

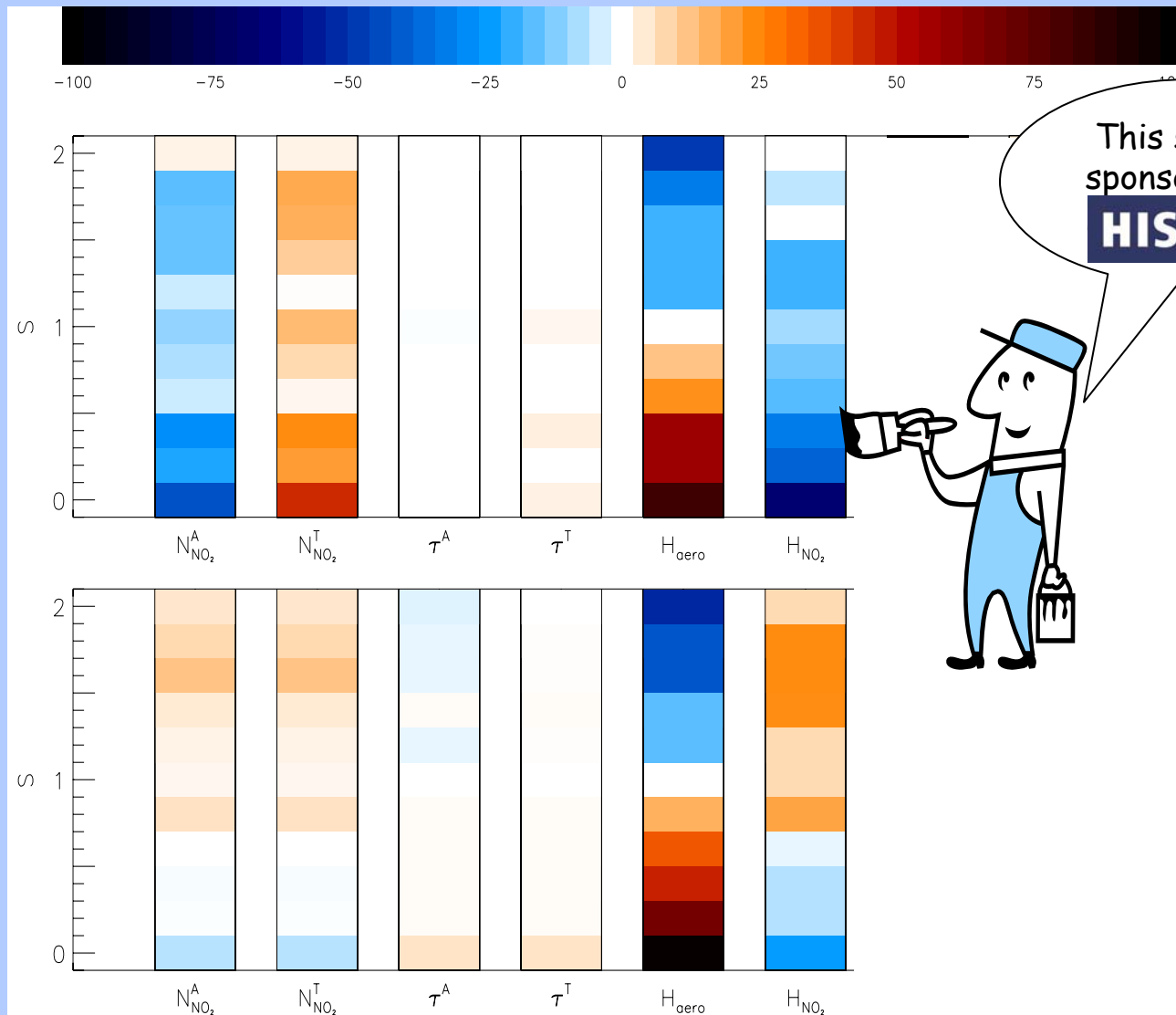
>> effect on model parameters



Sensitivity study: non-uniform boundary layer

sza = 40°, AOT = 0.2, H=1 km, 477 nm, NO₂ profile fixed

6 parameters

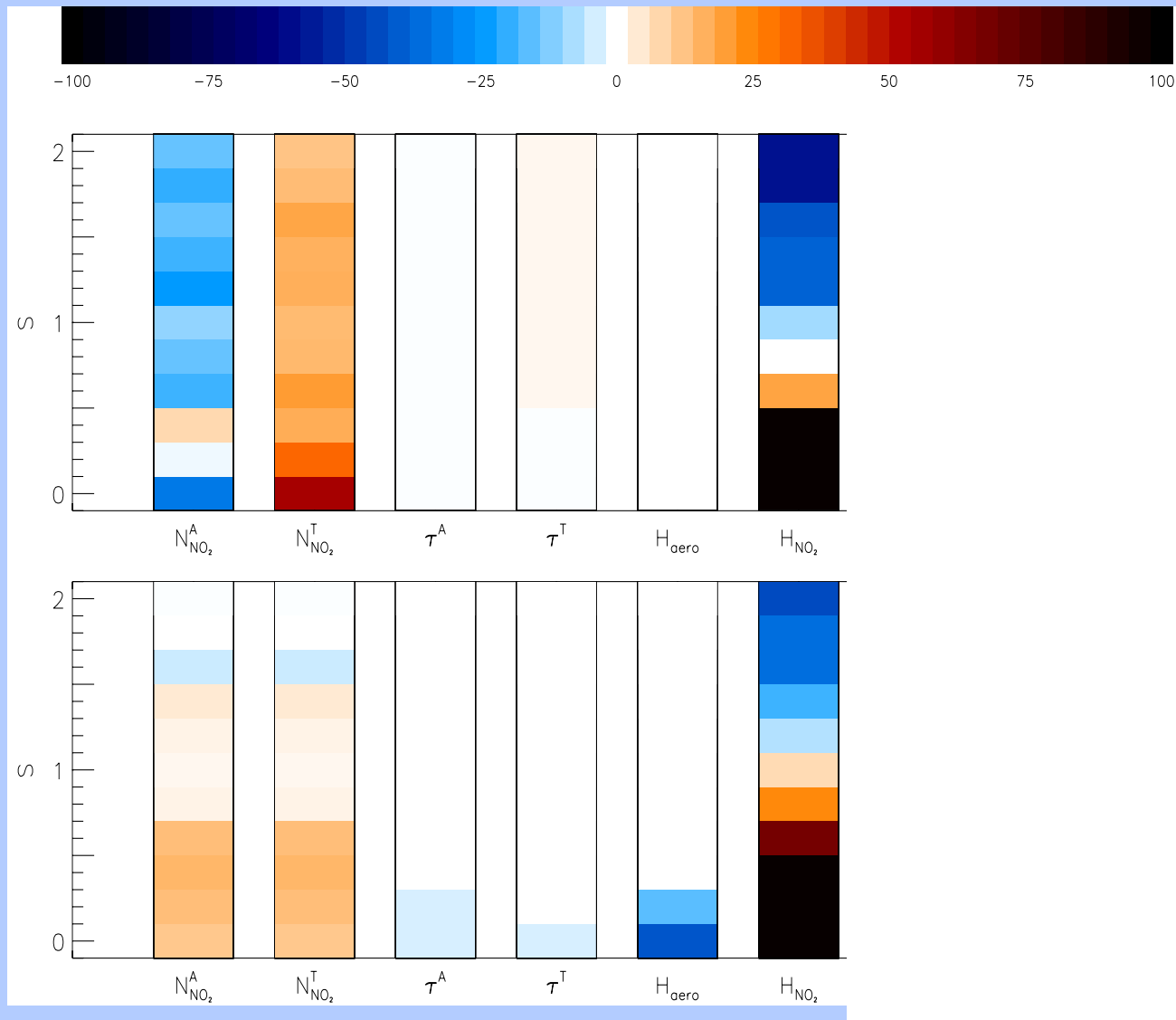


4 parameters

Sensitivity to non-uniform aerosol layer

sza = 40°, AOT = 0.2, H=1 km, 477 nm, Aerosol profile fixed

6 parameters

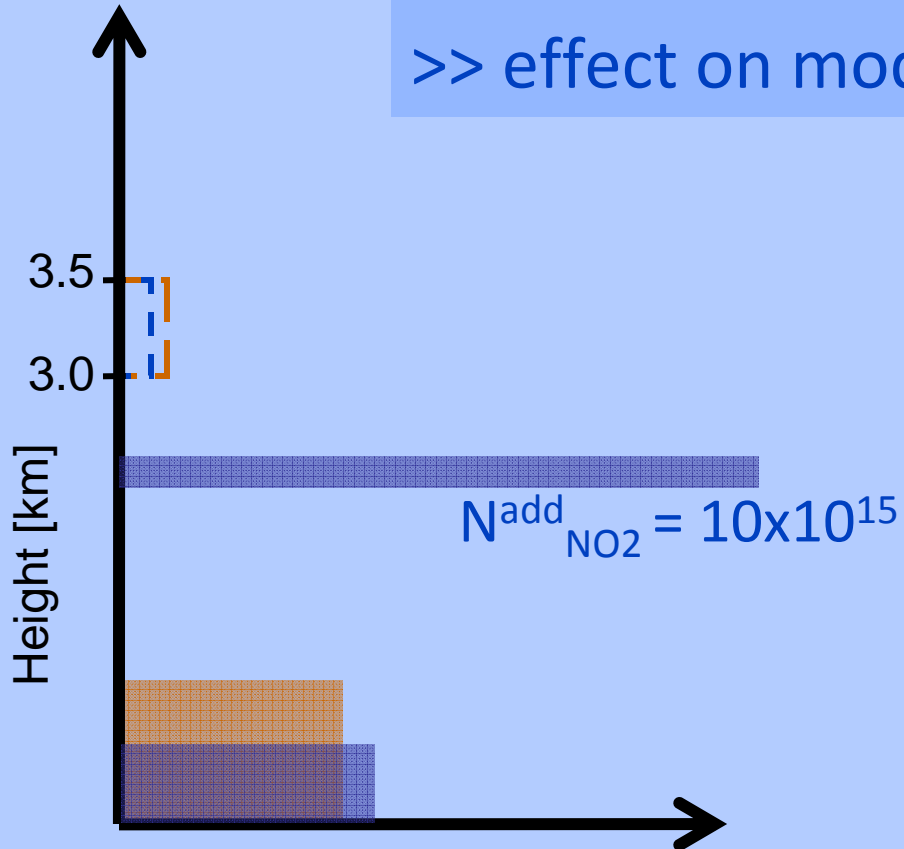


4 parameters

Sensitivity to non-uniform NO₂ layer

Sensitivity study #2 : free tropospheric NO_2 at different altitudes > 2 km

>> effect on model parameters



$N^{\text{A}}_{\text{NO}_2}$	15×10^{15}
$N^{\text{B}}_{\text{NO}_2}$	0
H_{NO_2}	0.4 km
H_{aer}	0.6 km
$\tau^{\text{A}}_{\text{aer}}$	0.2
$\tau^{\text{B}}_{\text{aer}}$	0

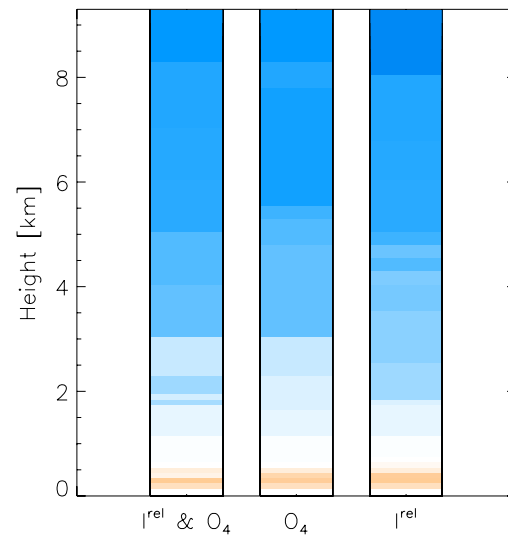
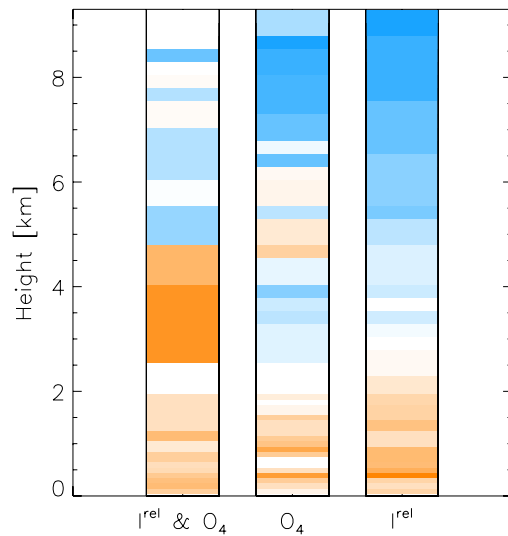
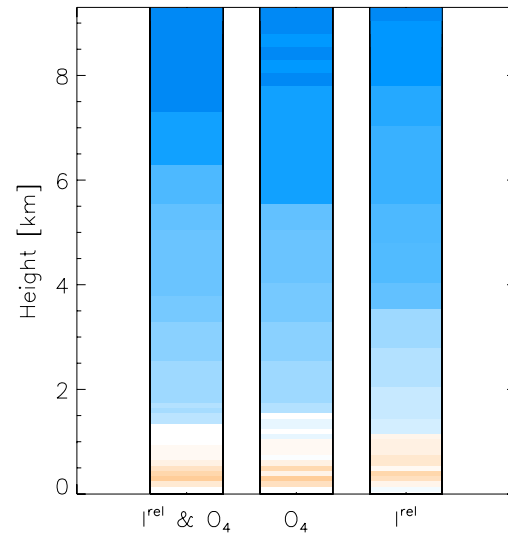
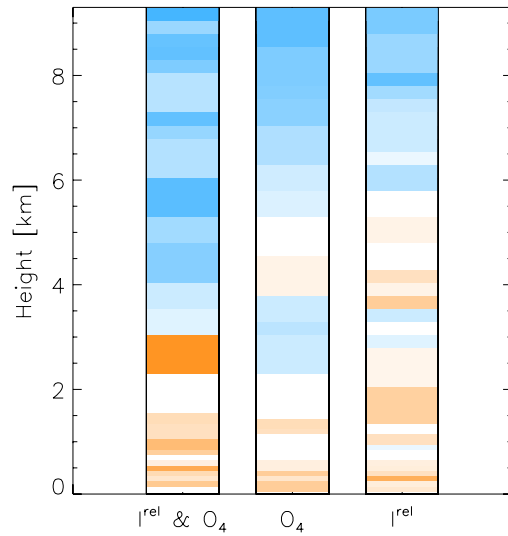
Sensitivity study: free tropospheric NO_2

SZA = 75

SZA = 40

6 parameters

4 parameters



Deviation in Tropospheric NO₂ column [%]

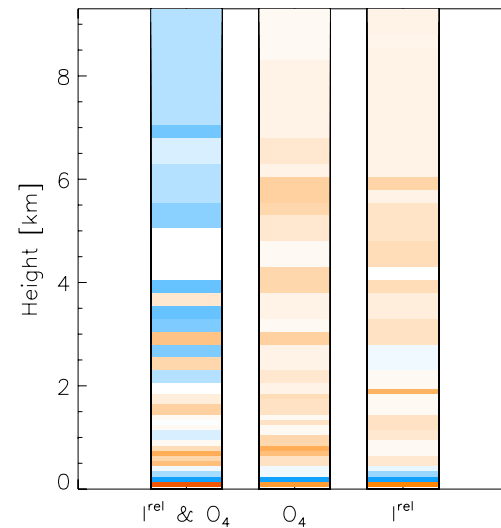
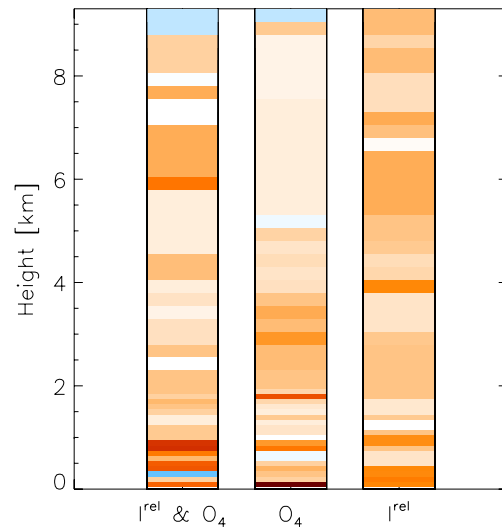
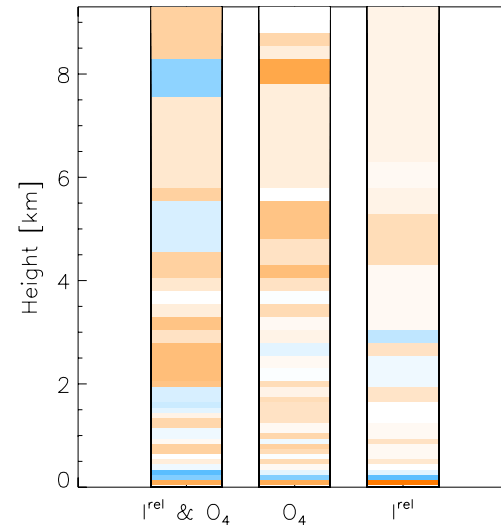
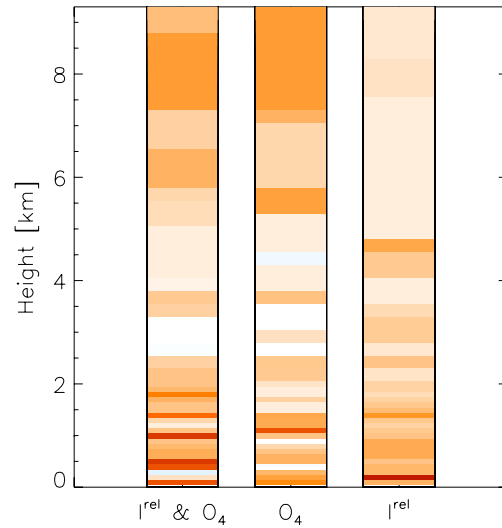
Sensitivity to elevated NO₂

SZA = 75

SZA = 40

6 parameters

4 parameters



Deviation in Tropospheric NO₂ column [%]

Sensitivity to elevated aerosol

Conclusions Sensitivity Study

The 6P model is somewhat better capable of retrieving high NO₂, better accuracy than 4P, but low precision

The 6P model however is also more sensitive to inhomogeneities in the lowest NO₂ layer, leading to larger errors in the total column than in the 4P case

The second aerosol layer in the 6P model in general leads to an overestimated NO₂ column

Did we choose the best set of parameters?

The need for ground based column observations

The MAX-DOAS technique

Development of a retrieval model

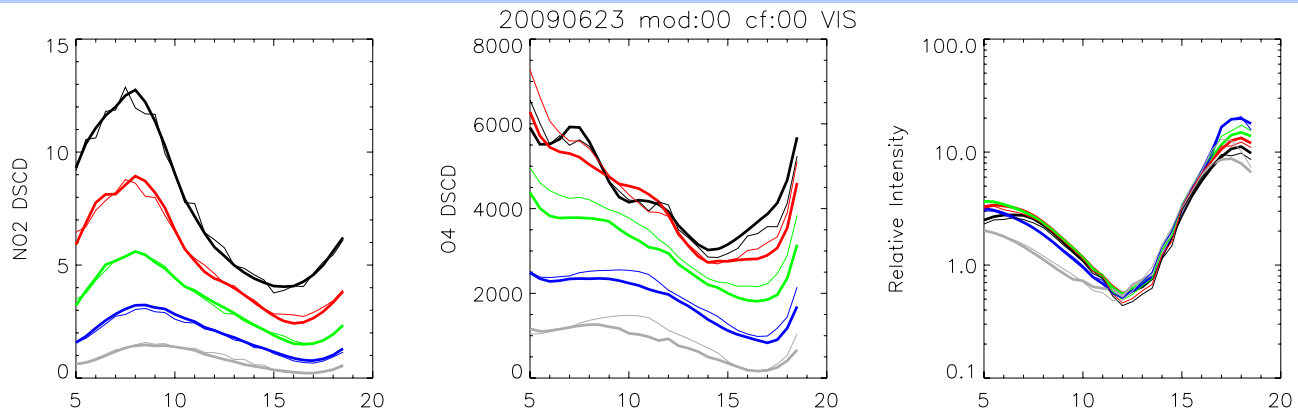
Sensitivities and error analysis

Comparisons

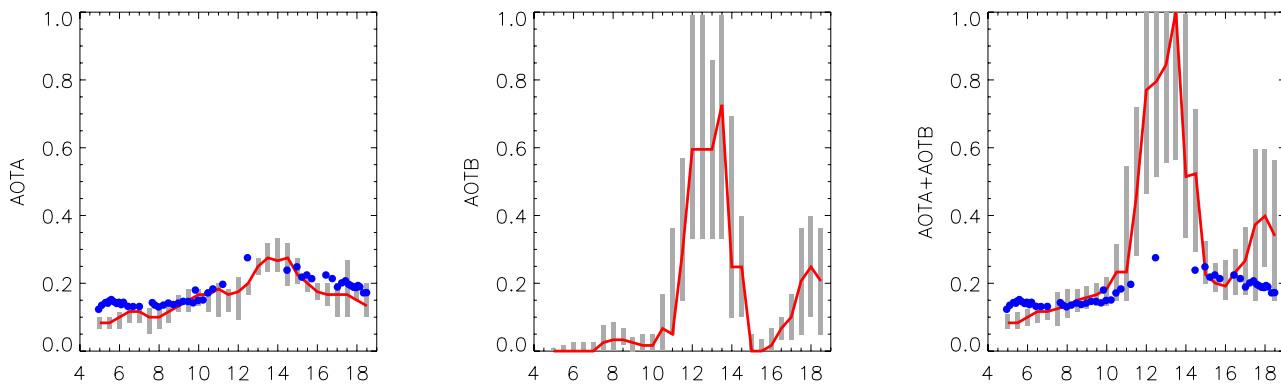
Summary and Conclusions

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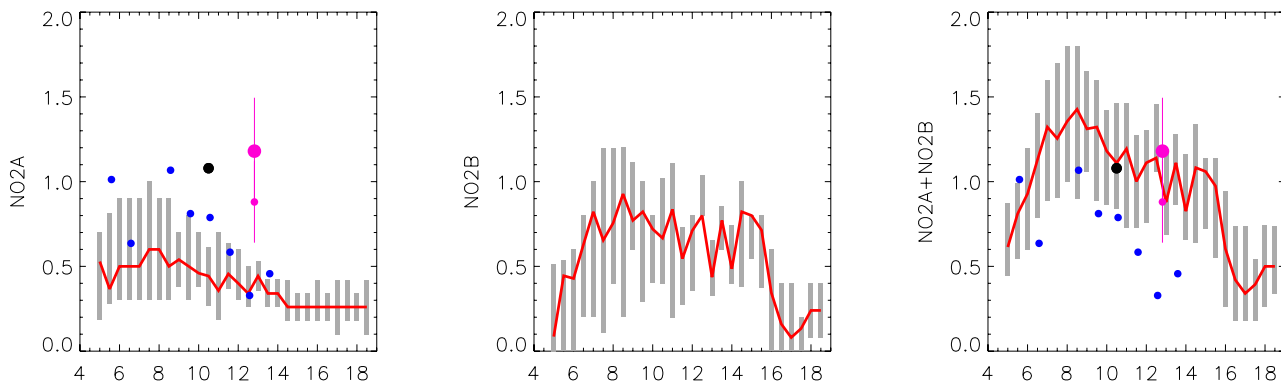
Measurements



AOT retrieval



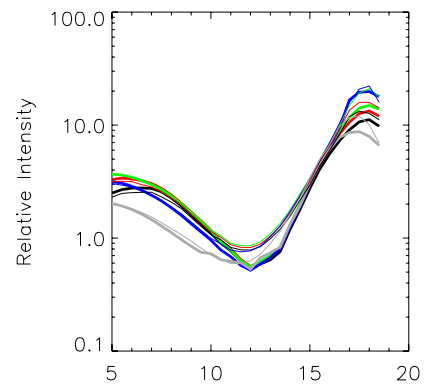
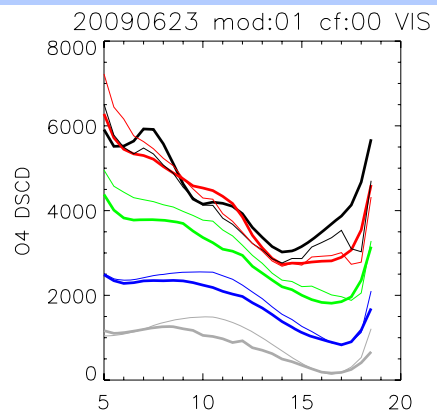
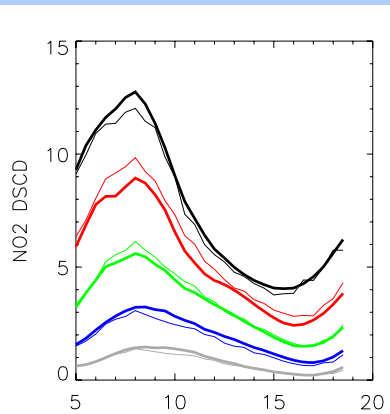
NO2 retrieval



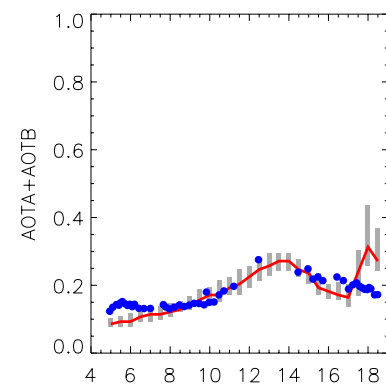
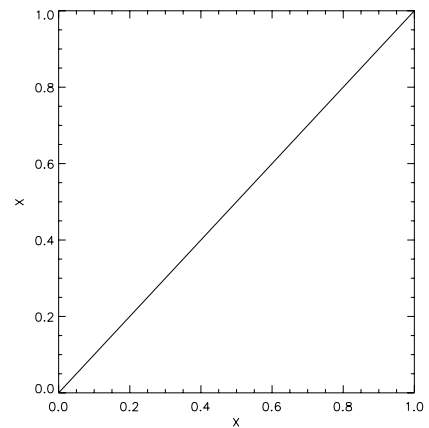
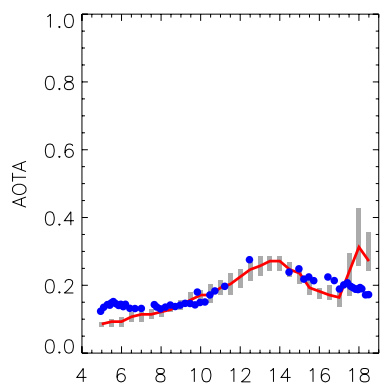
• AERONET

• Lidar RIVM

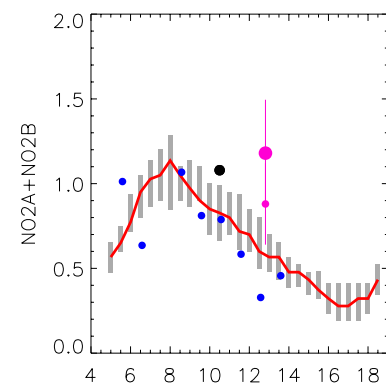
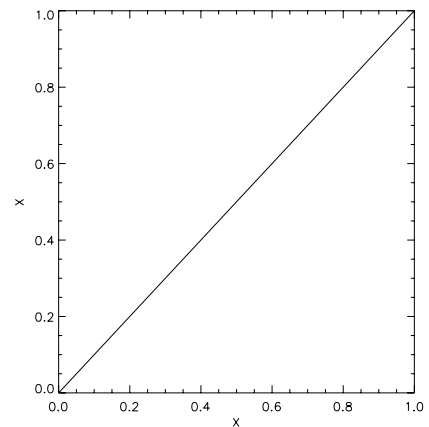
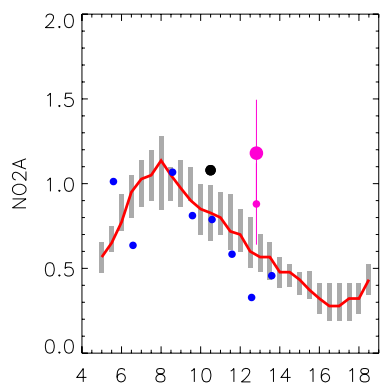
Measurements

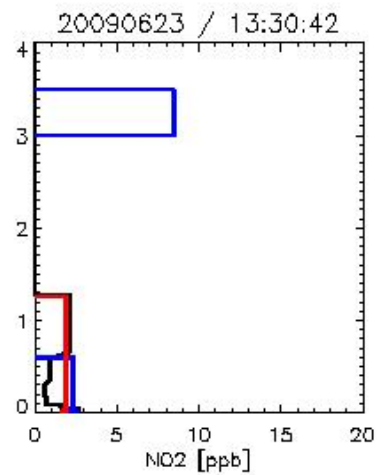
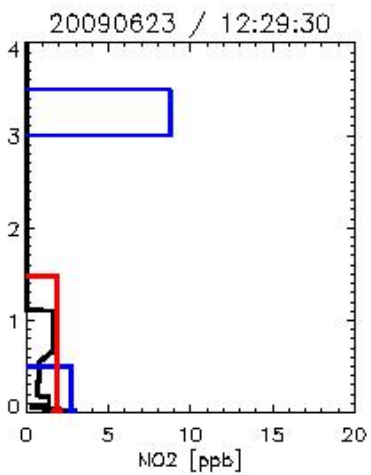
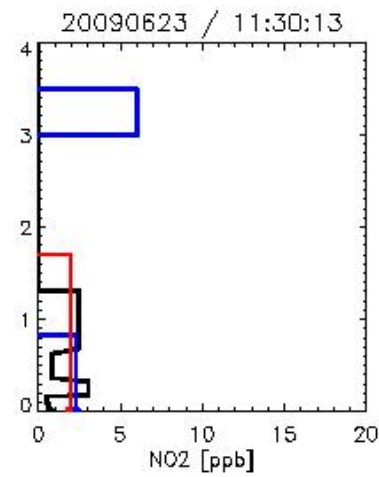
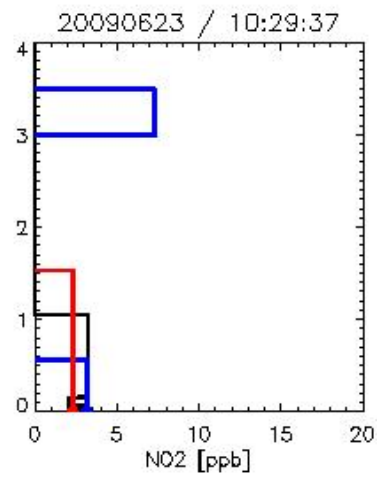
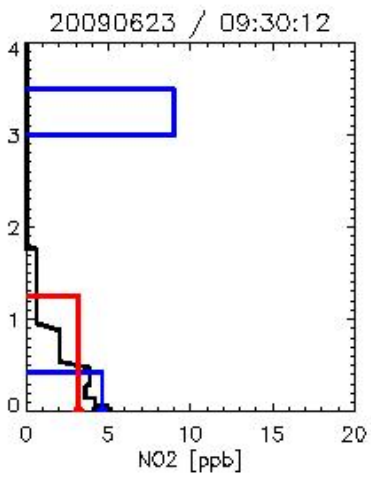
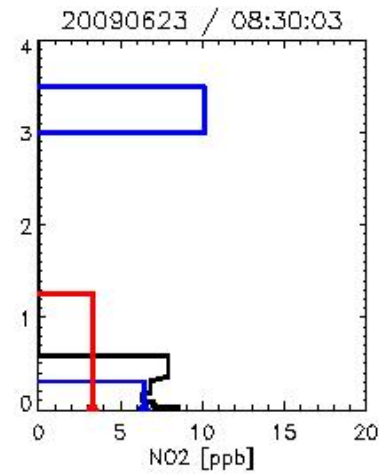
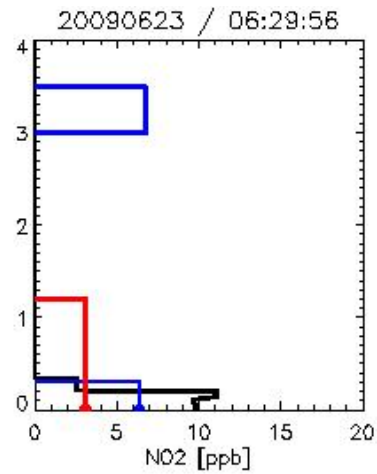
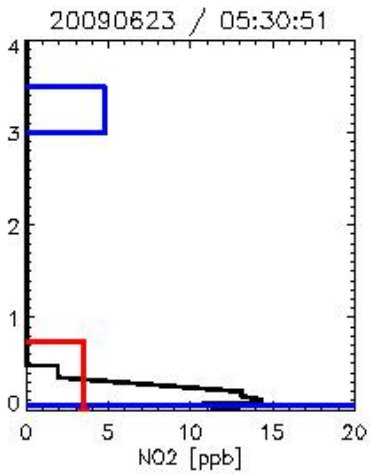


AOT retrieval



NO2 retrieval

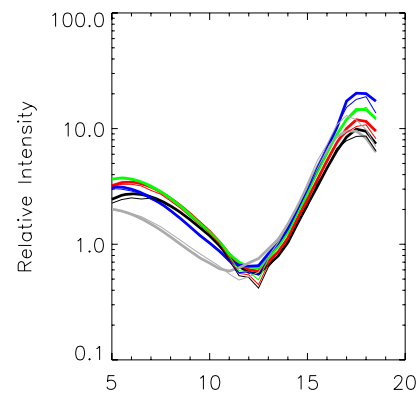
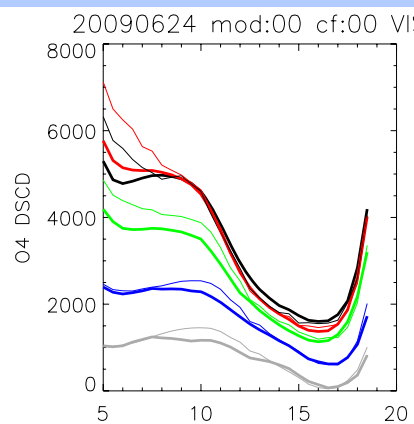
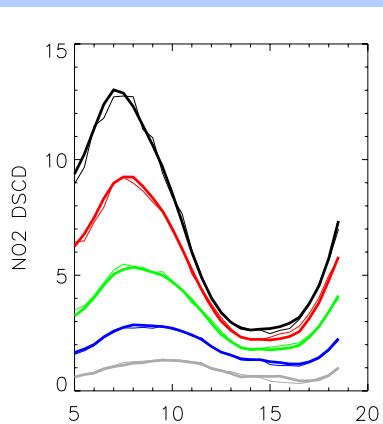




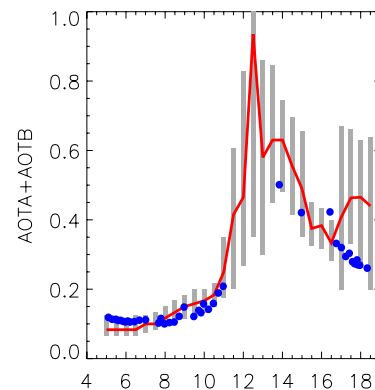
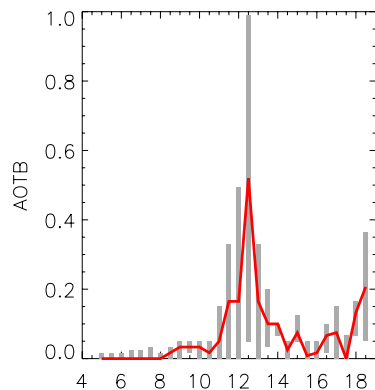
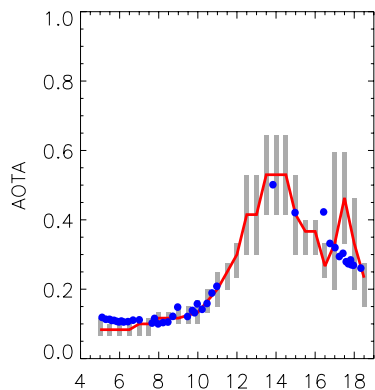
- RIVM Lidar
- MAX-DOAS 4P
- MAX-DOAS 6P

Courtesy lidar data: S. Berkhout, RIVM

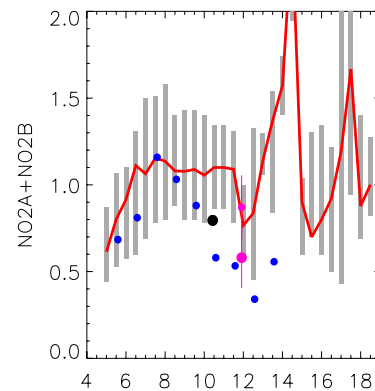
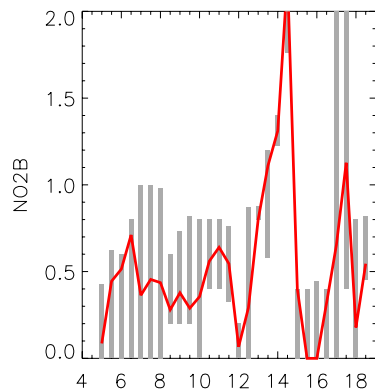
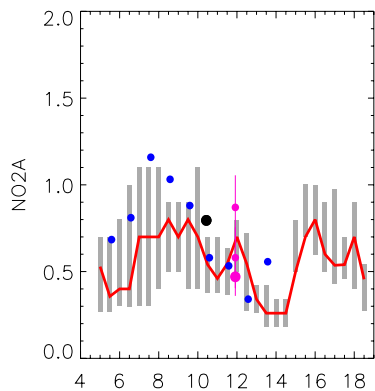
Measurements



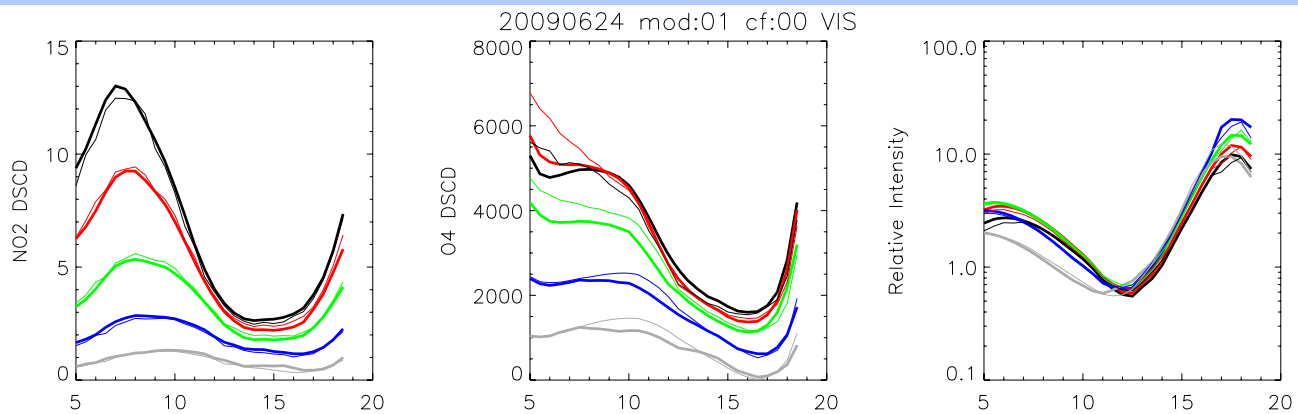
AOT retrieval



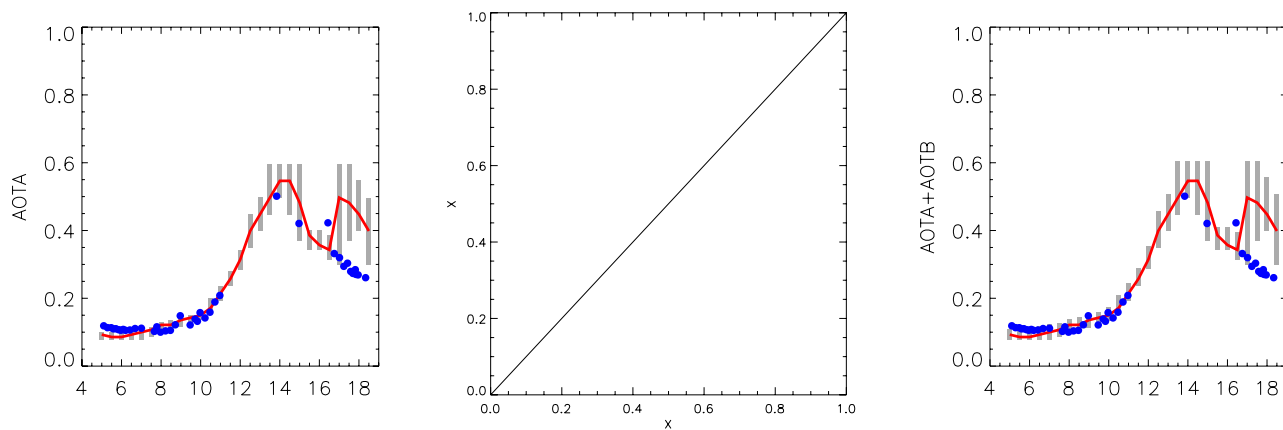
NO2 retrieval



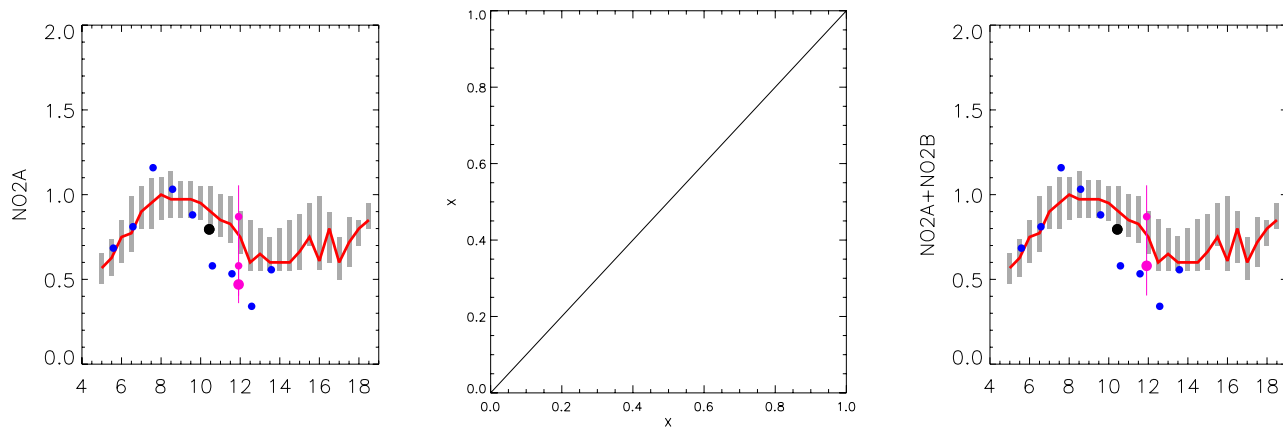
Measurements

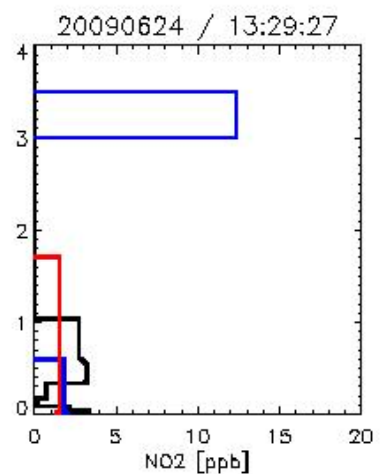
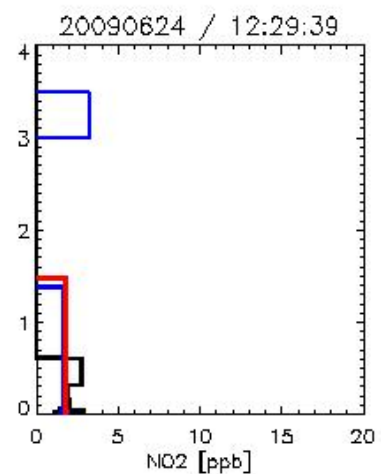
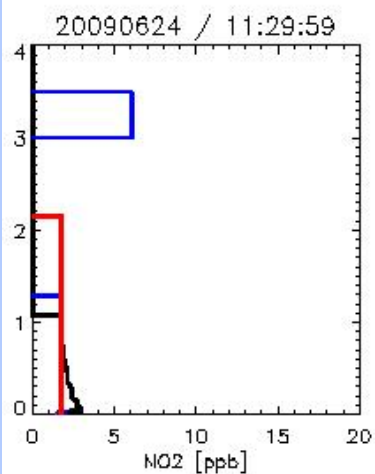
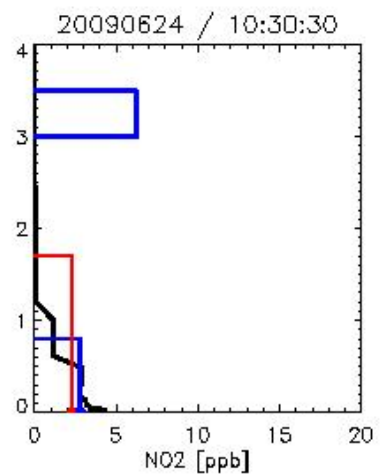
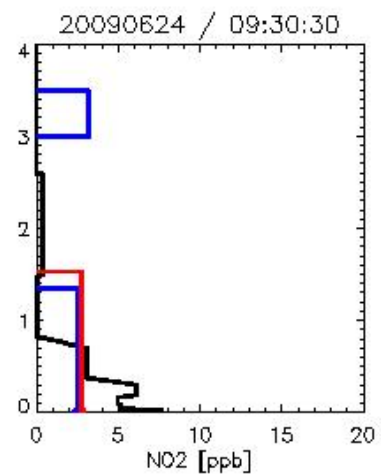
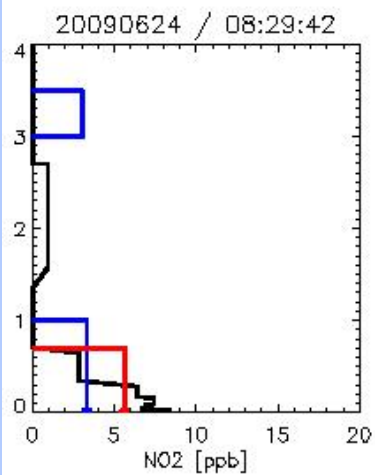
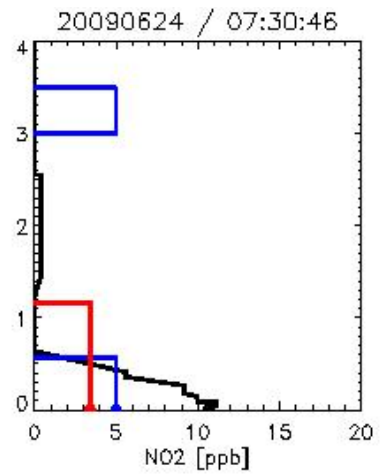
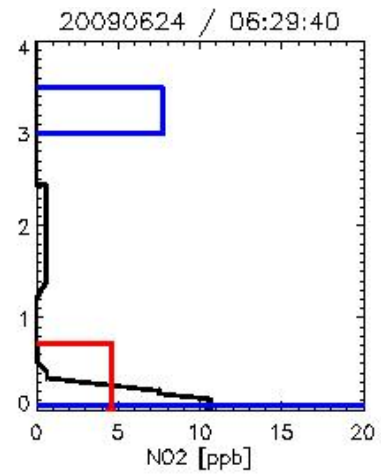
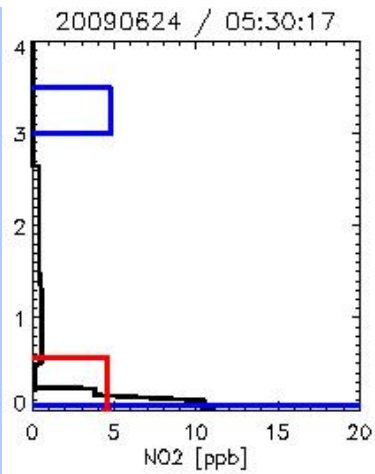


AOT retrieval

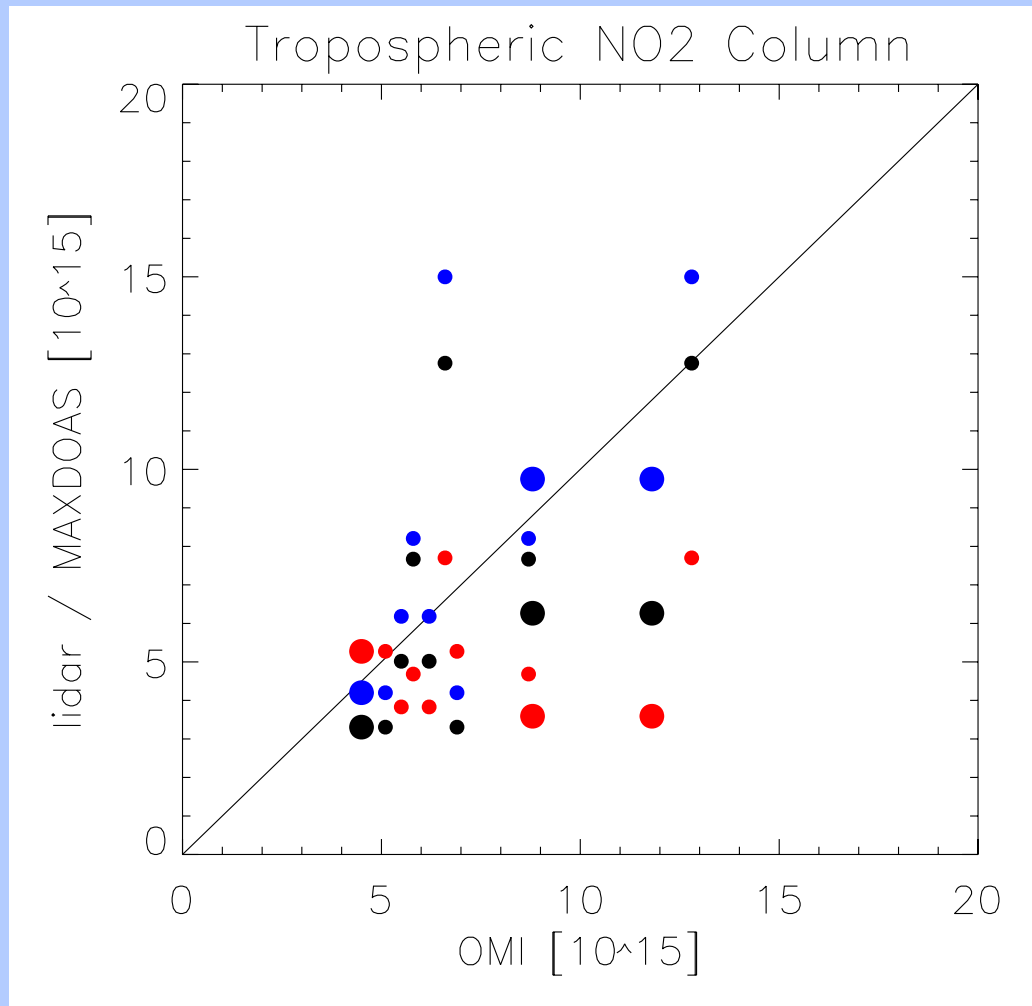


NO2 retrieval





- RIVM Lidar
- MAX-DOAS 4P
- MAX-DOAS 6P



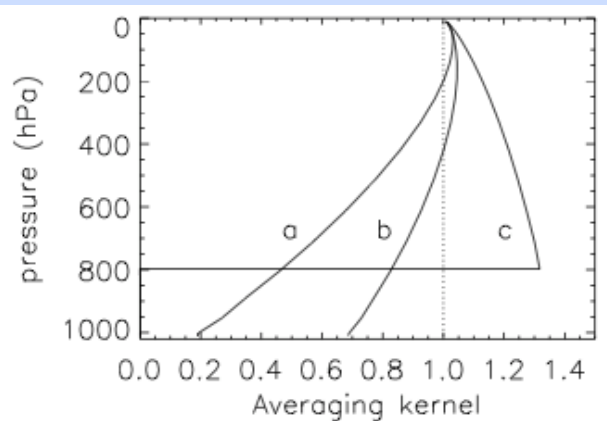
- Lidar
- MAX-DOAS 4P
- MAX-DOAS 6P

All points: OMI pixel center < 15 km from Cabauw
 Enlarged points: OMI pixel center < 10 km from Cabauw **and** small pixels

Characteristics of NO₂ profile shape determining the accuracy of the tropospheric NO₂ column / profile retrieval

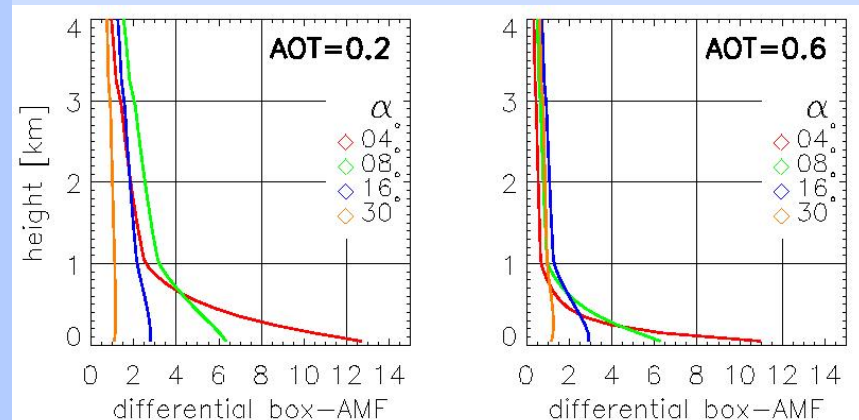
OMI

- 1 Ratio BL / FT
- 2 Height of FT NO₂
- 3 Height of BL
- 4 NO₂ profile shape BL



MAX-DOAS

- 1 NO₂ profile shape BL
- 2 Aerosol profile shape BL
- 3 Height of BL
- 4 Ratio BL / FT
- 5 Height of FT NO₂



The need for ground based column observations

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Summary and Conclusions

MAX-DOAS can retrieve tropospheric columns using a high elevation that is insensitive to the profile shape of aerosols and NO₂

Detailed satellite validation requires knowledge of vertical profile

Attempts to separate free tropospheric- from boundary layer NO₂ give either large uncertainties (6P) or negative biases (4P, FT NO₂)

