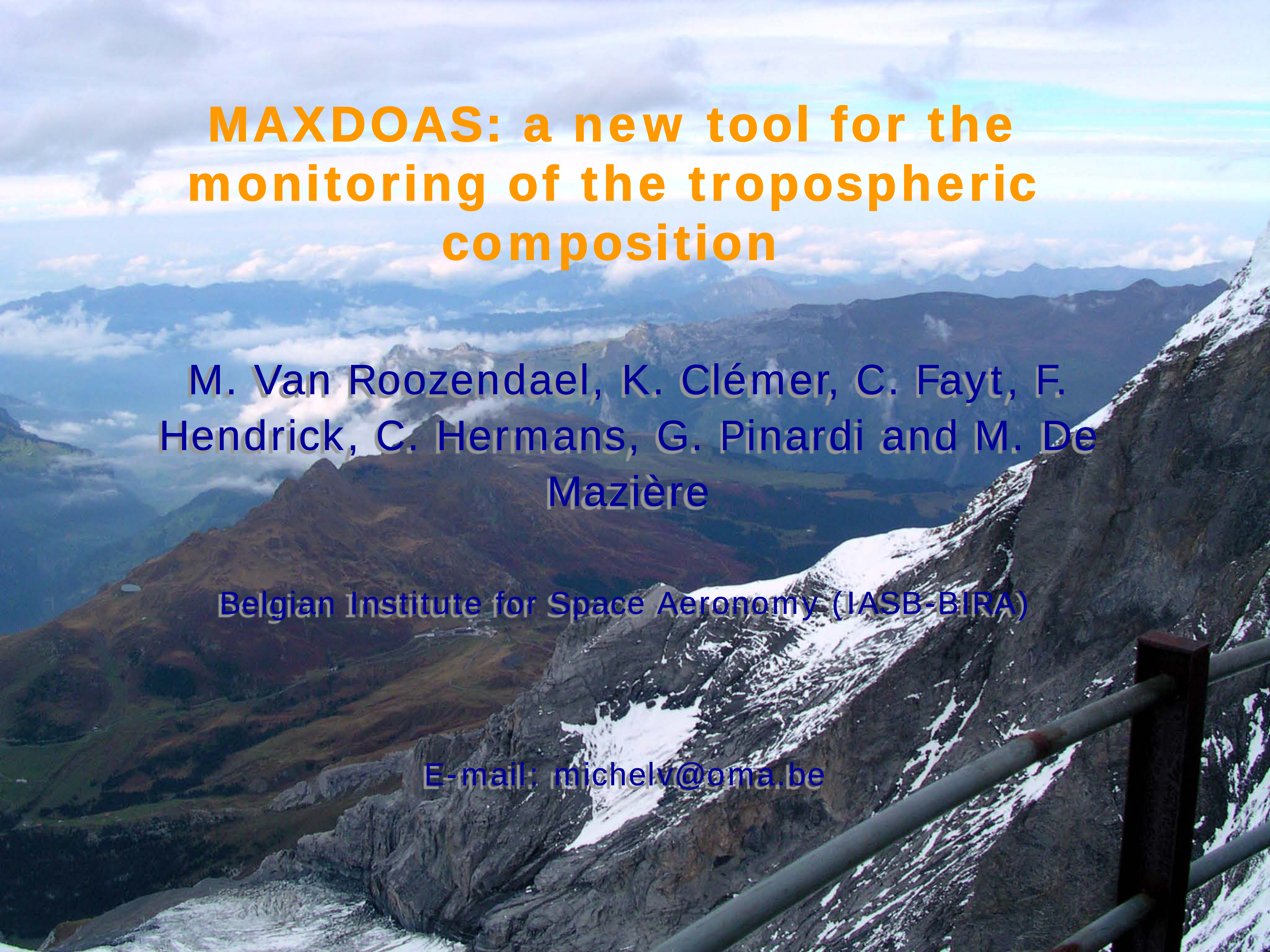

MAXDOAS: a new tool for the monitoring of the tropospheric composition

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Content

- Introduction : principle of DOAS and MAXDOAS
- The BIRA-IASB MAXDOAS system
- Tropospheric column retrieval using the geometrical approximation
- Progress towards aerosol and trace gas profiles retrieval using an Optimal Estimation inversion scheme
- Conclusions and perspectives for implementation at Jungfraujoch

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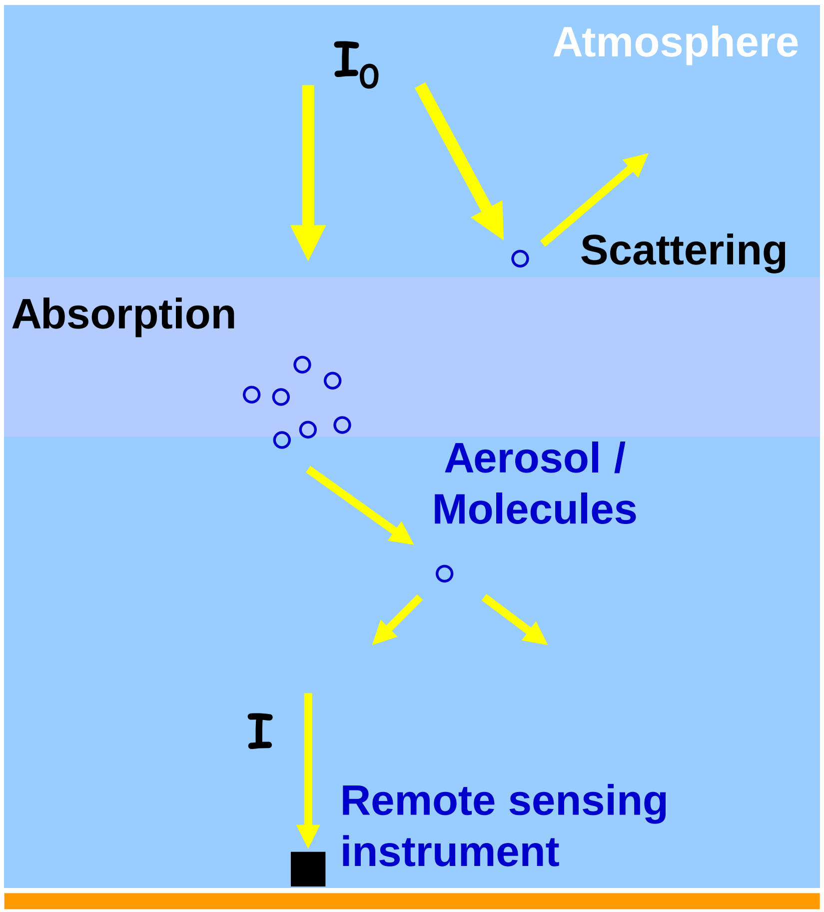
Remote sensing in UV-Vis region

- Solar radiation is attenuated when traveling through the atmosphere, following the Beer-Lambert law:



$$I(\lambda) = I_0(\lambda)\exp\left[-\sum_i SCD_i.\sigma_i(\lambda) - \tau_{diffusion}(\lambda)\right]$$

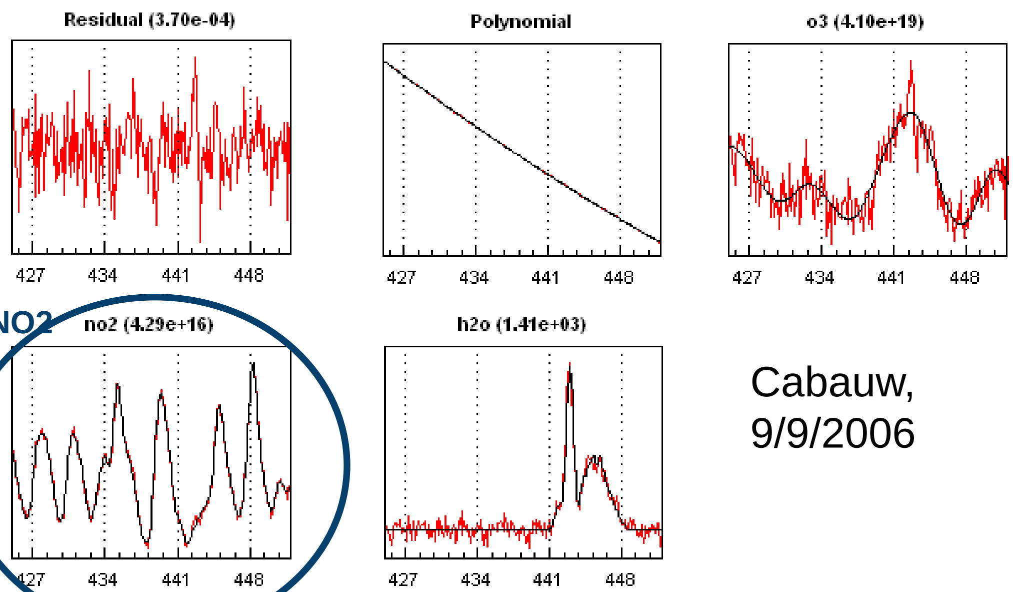
σ_i : molecular absorption cross-section
SCD_i: slant column density
$\tau_{diffusion}$: Rayleigh and Mie scattering

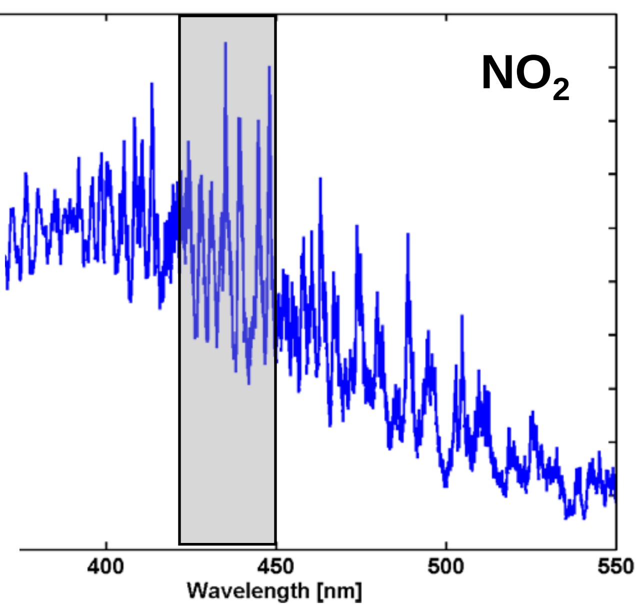
Principle of DOAS

- Use high frequency absorption structures to identify absorbers
- Quantify abundance based on measure of the differential absorption (i.e. difference between strongly and weakly absorbing wavelengths)

$$\ln \frac{I(\lambda)}{I_0(\lambda)} = -\sum_i SCD_i.\sigma_i(\lambda) - P_\lambda$$

SCD_{NO2}

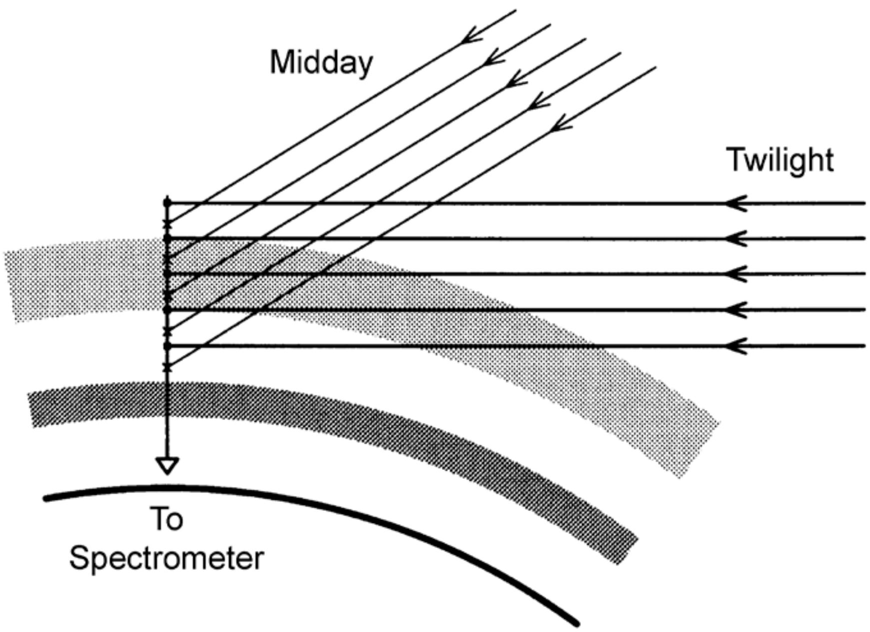




Cabauw, 9/9/2006

Geometry (1) Zenith-sky twilight

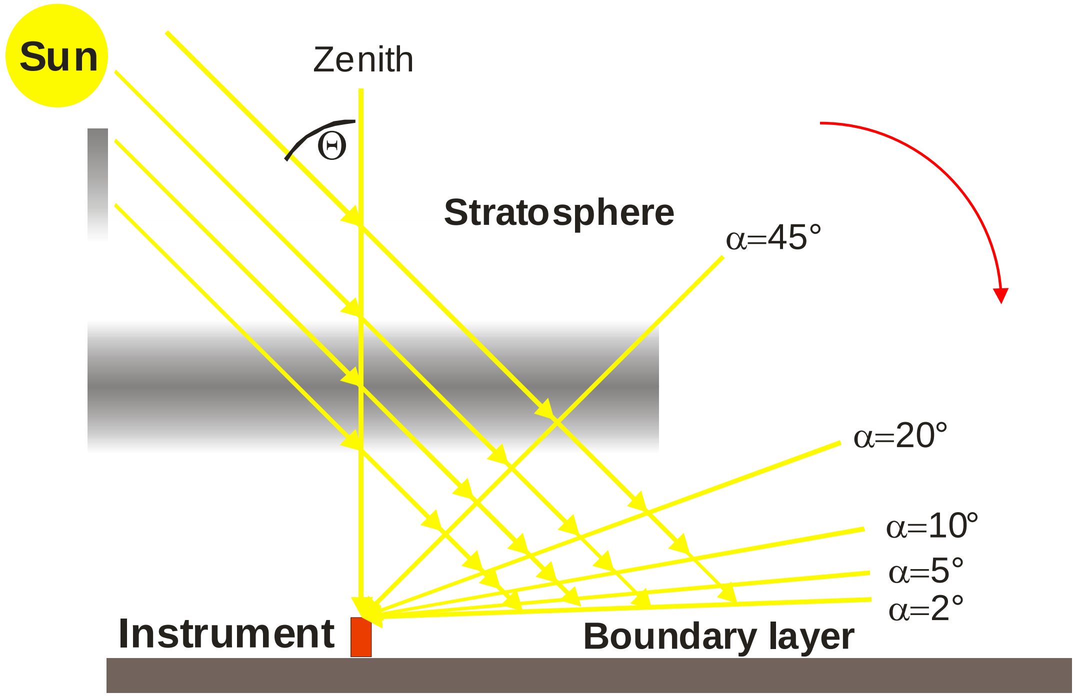
- Large geometrical enhancement factors at twilight



- High sensitivity to stratospheric trace gases (O_3, NO_2, BrO, ...)
- Weak contamination by tropospheric absorbers
- Ideal for stratospheric monitoring (e.g. SAOZ)

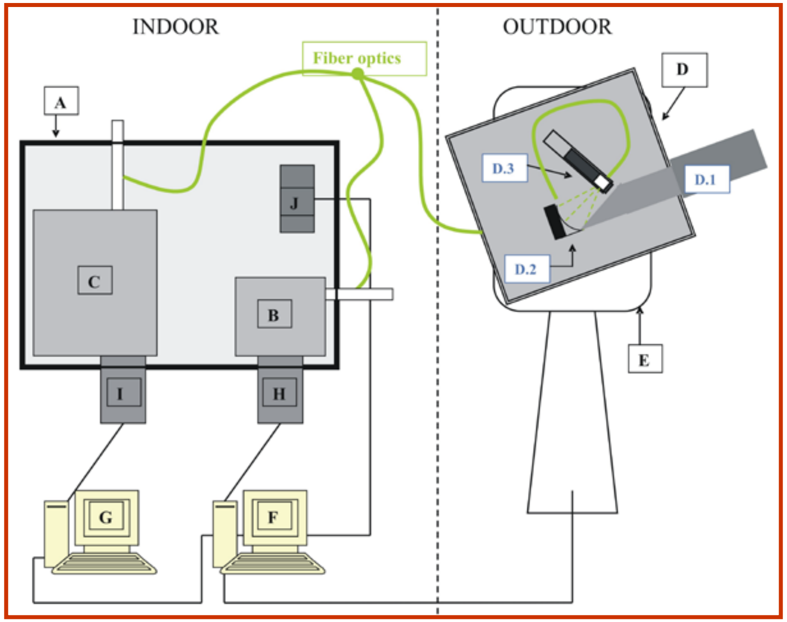
Geometry (2) MAXDOAS

- **Multi-Axis DOAS** (MAXDOAS)
  → zenith + scan of multiple elevation angles down to horizon



- Increases sensitivity to lower troposphere
- Provides information on the vertical distribution of gases and aerosols in the troposphere
- Retrieval complicated !

BIRA-IASB MAXDOAS system



Main parts:

- sun tracker
- optical head
- 2 spectrometers
- 2 CCD
- 2 computers

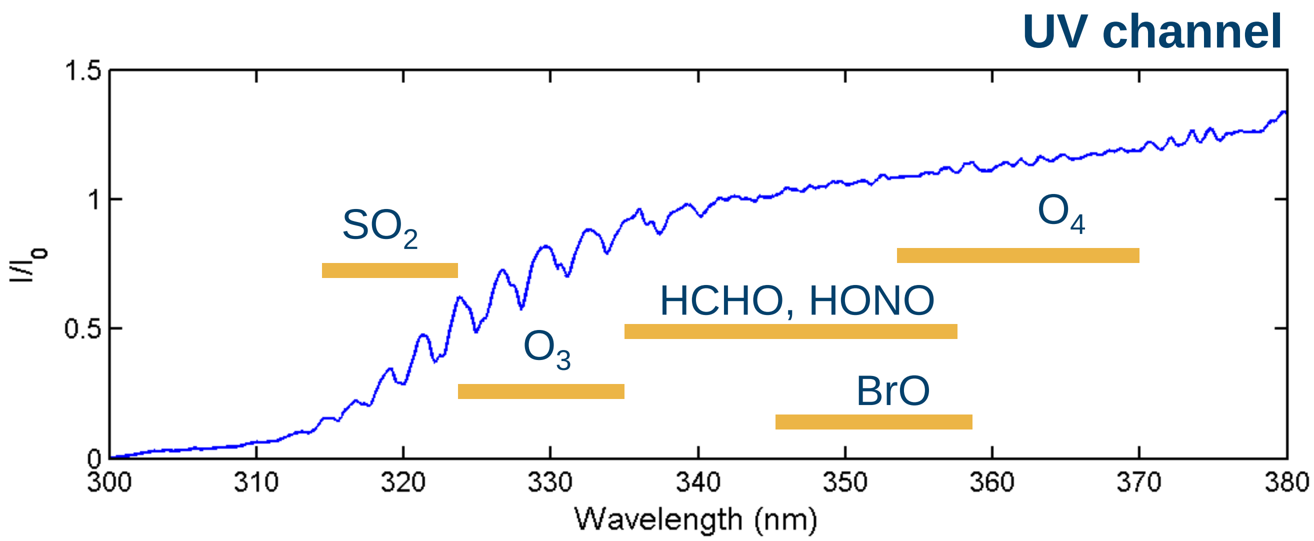
2 Channels:

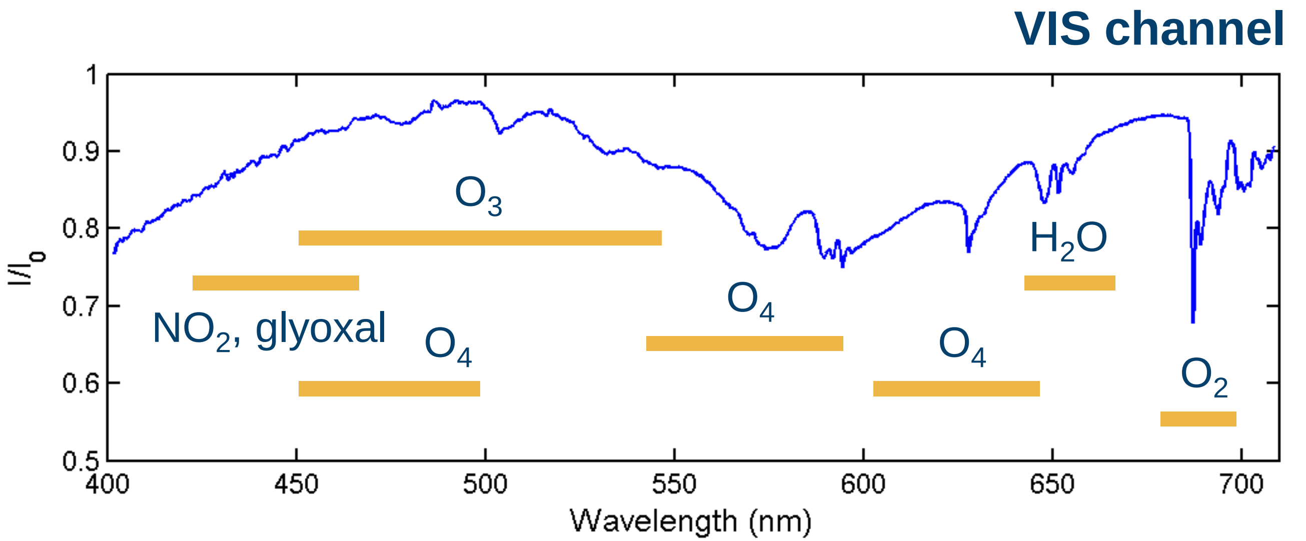
- UV = 300 - 390 nm
- VIS = 400 - 720 nm

Measurement modes:

- Scattered light: horizon to zenith
- Direct sun-measurement

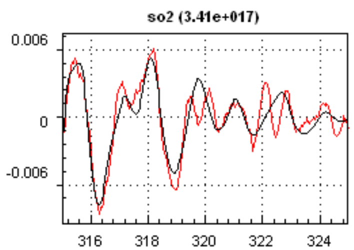
Spectral intervals in UV and VIS channels



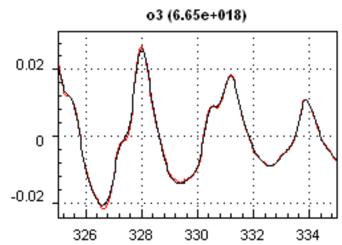


Sample DOAS fits for target trace gases

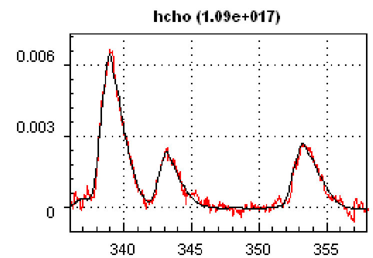
SO_2



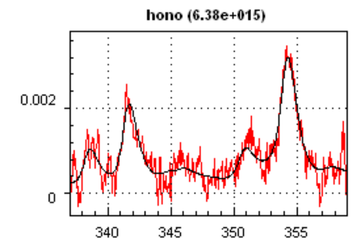
O_3 (Huggins)



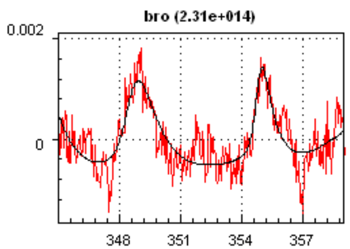
HCHO



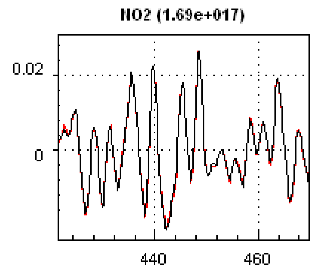
HONO



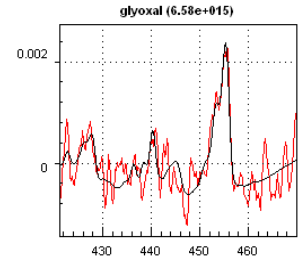
BrO



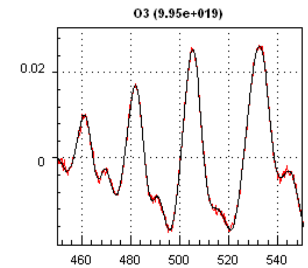
NO_2



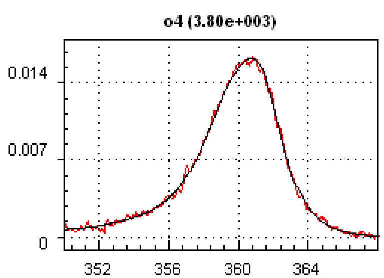
Glyoxal



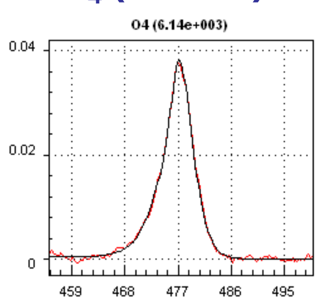
O_3 (Chappuis)



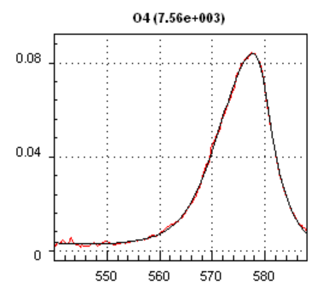
O_4 (360 nm)



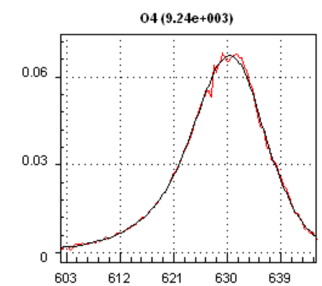
O_4 (477 nm)



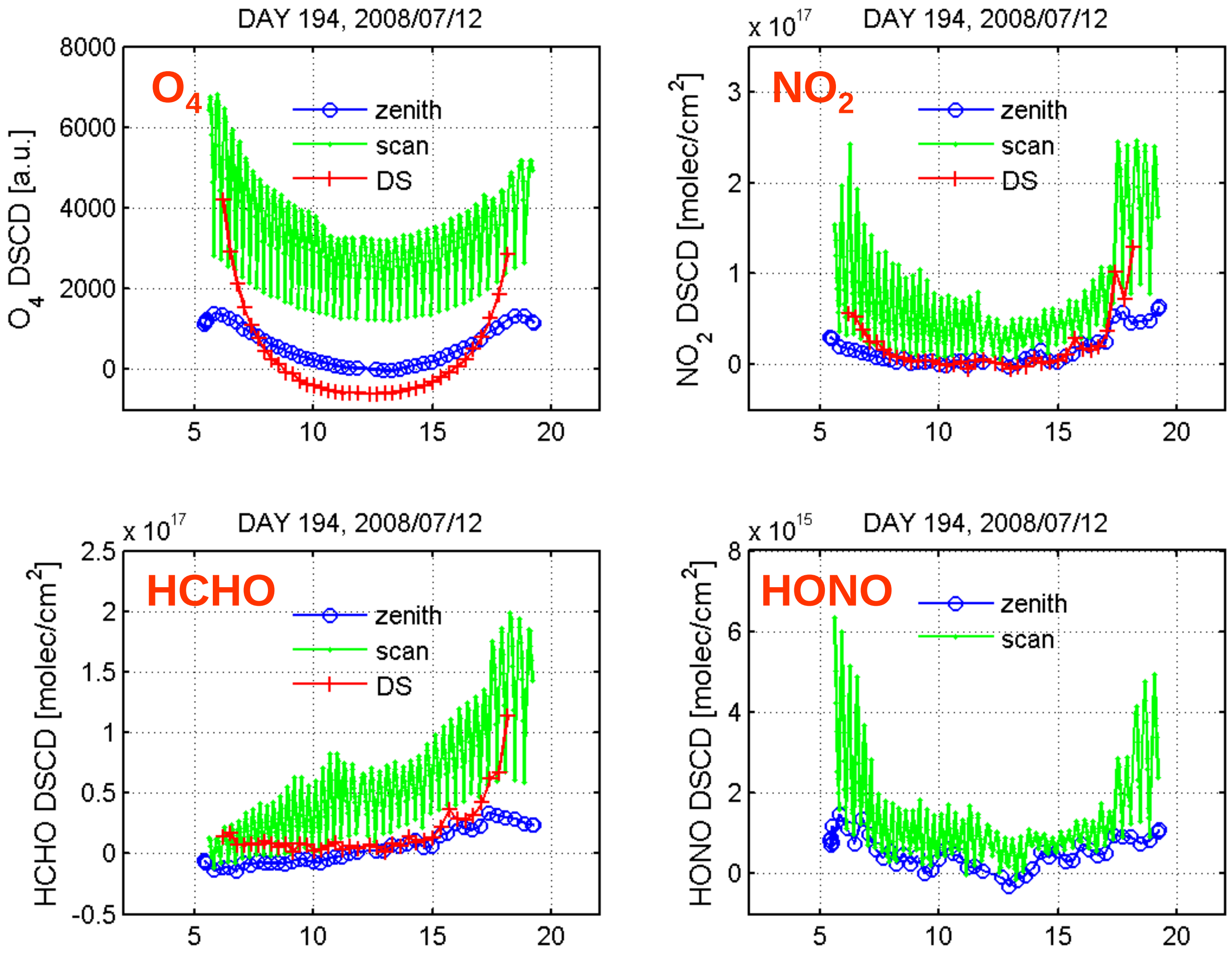
O_4 (577 nm)



O_4 (630 nm)

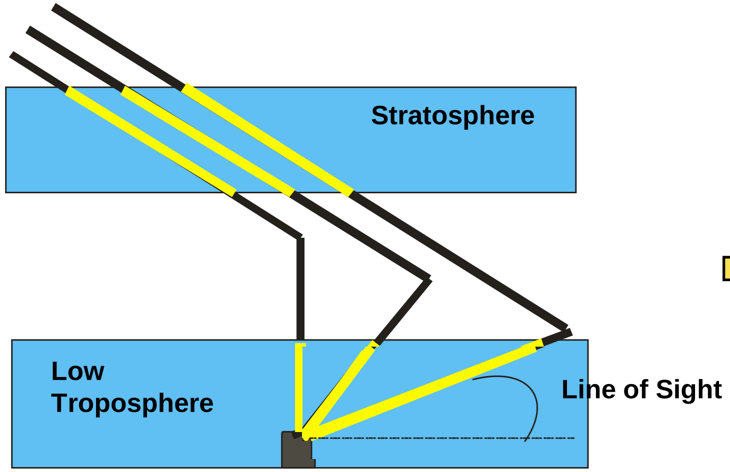


Example of raw slant column measurement



MAXDOAS tropospheric retrievals

- The easy case ➔ tropospheric column retrieval using the geometrical approximation



$$\Rightarrow \quad VC_{trop} = \frac{SC_{off-axis} - SC_{zenith}}{\sin^{-1}(LOS) - 1}$$

- If one can assume that the gas layer is confined below the last scattering point, the vertical column can be derived from simple geometrical considerations
- Valid for moderate LOS angles (15° - 30°)
- Provides only tropospheric column (no profile information)

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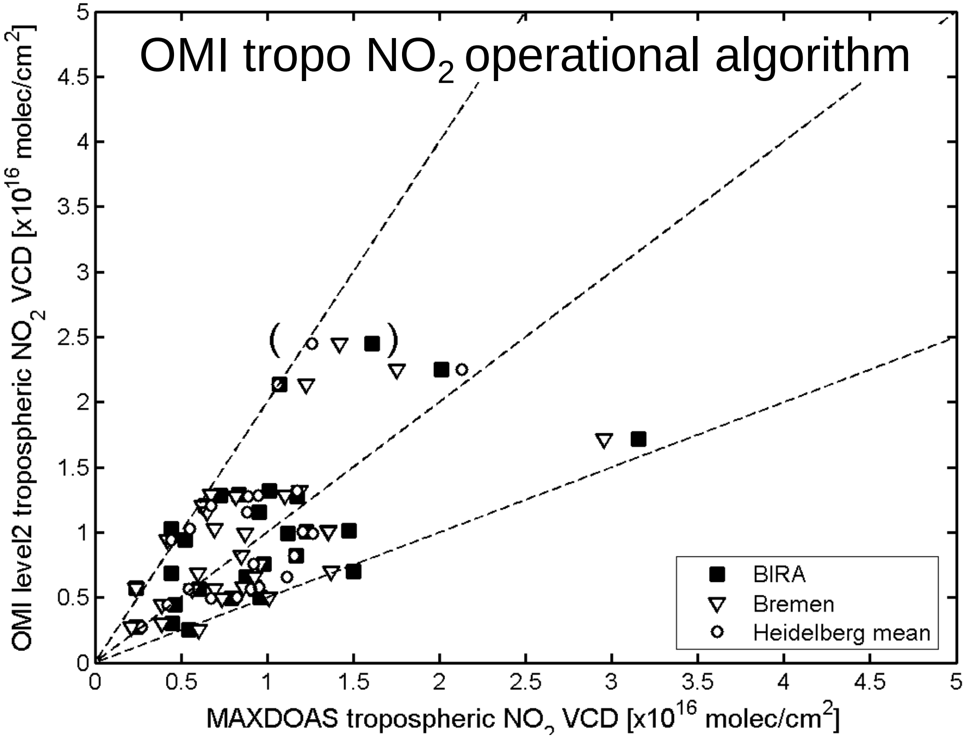
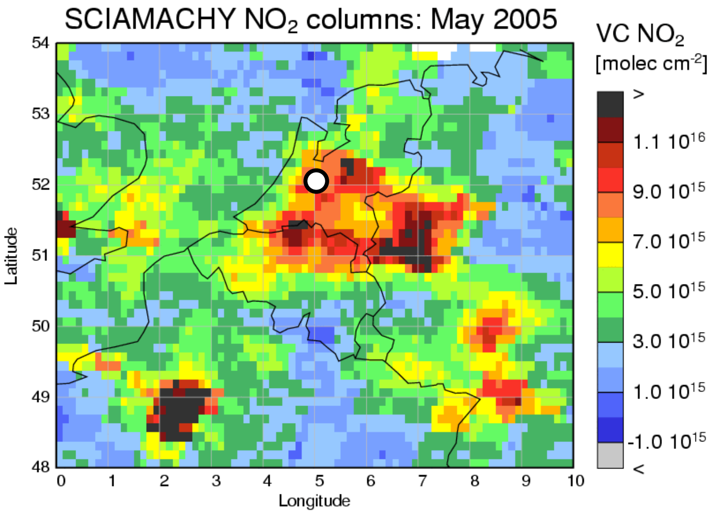
Application: satellite validation

E.g. SCIAMACHY and OMI tropospheric NO_2 validation during DANDELIONS campaigns (summer 2005 and autumn 2006 in Cabauw, NL)





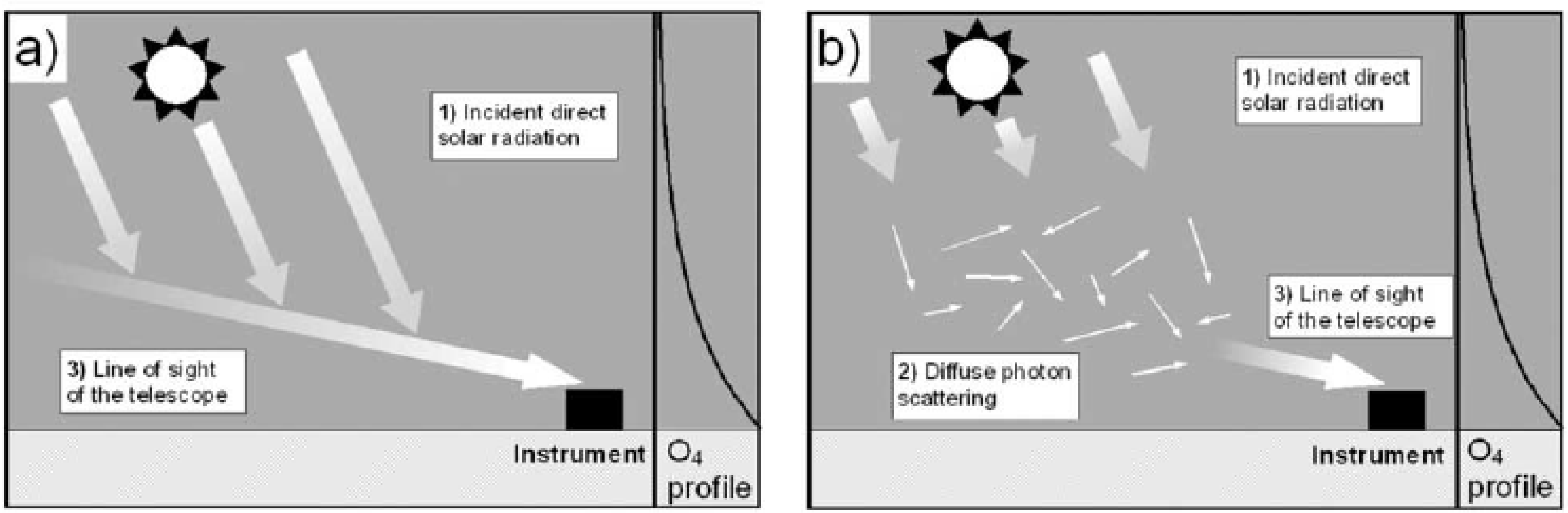
Brinksma et al., JGR, 2008

Aerosol and trace gas profile retrieval

- Definition SCD: $$SCD = \int_0^L \rho(s)ds$$

- In the case of O_4, ρ(z) is well-known and nearly constant (it varies with the square of molecular O_2)

➔ Changes in the observed O_4 DSCD are due to changes in the light path distribution.



Wagner, JGR, 2004
Friess, JGR, 2006

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Aerosol information in O_4 DSCD

The length of the light path through the atmosphere and thus the observed SCD of a trace gas will depend on the vertical distribution and optical properties of the aerosol present in the atmosphere

Measurements used for aerosol properties retrievals:

- O_4 DSCD ($SCD_{off\text{-}axis}$-SCD_{zenith}) and DI ($DI_{off\text{-}axis}/DI_{zenith}$)
- @ 4 wavelengths
- For different viewing geometries (SZA, relative azimuth angle, elevation angle)

Forward model

- Calculate radiances and slant columns @ different wavelengths and viewing geometries for a certain atmosphere.

  ↳ *INPUTS: - P, T*
  - Surface albedo
  - Trace gases (σ,ρ)
  - Aerosol (extinction profile
  single scattering albedo
  phase function)

- **Forward model** ➔ Linearized discrete ordinate radiative transfer code (LIDORT v3.3) *(Spurr, 2007)*

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Inversion method

- Optimal Estimation method *(Rodgers, 2000)*

$$k_{i+1} = k_i + (S_a^{-1} + K_i^T S_\varepsilon^{-1} K_i)^{-1}[K_i^T S_\varepsilon^{-1}(y - F(k_i)) - S_a^{-1}(k_i - k_a)]$$

k = aerosol extinction vertical profile

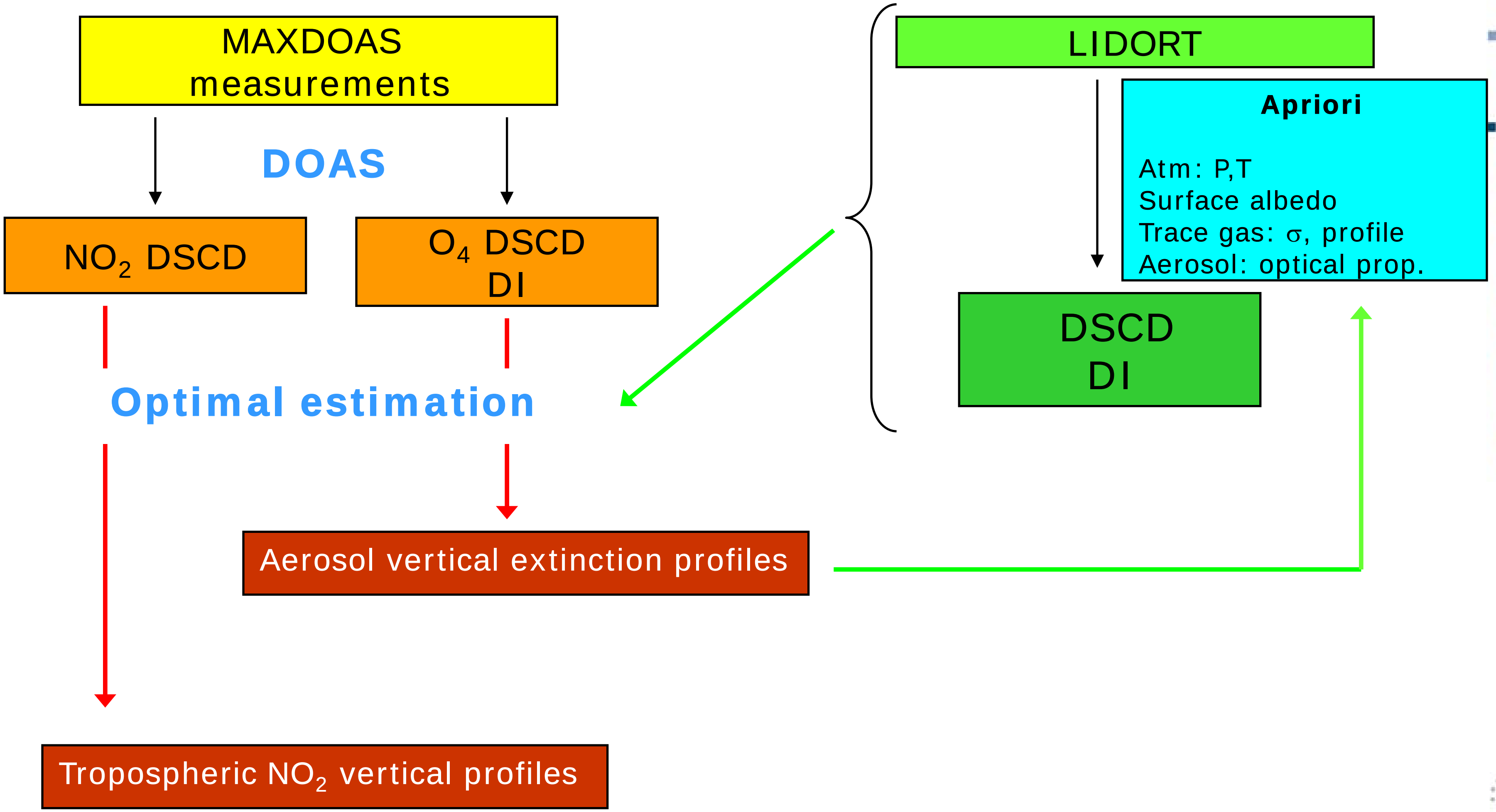
k_a= apriori aerosol extinction vertical profile
S_a = uncertainty covariance matrix of the apriori profile

F = Forward model (LIDORT)
y = measurement (O_4 DSCD and/or DI)
S_ε = uncertainty covariance matrix of the measurement

K = weighting functions = $\partial \mathbf{y}/\partial \mathbf{k}$

Advantage of LIDORT : analytical calculation of the weighting functions

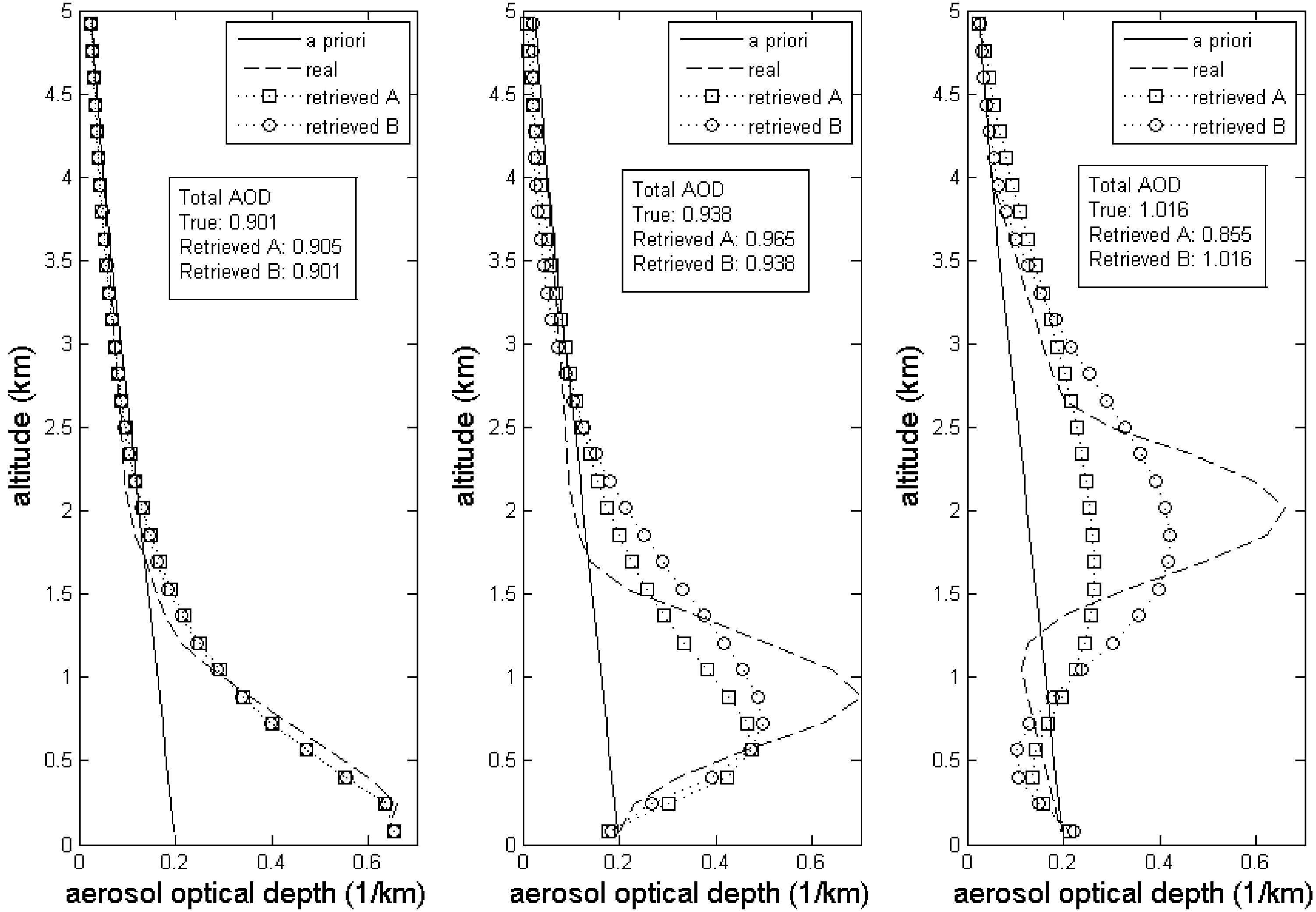
MAXDOAS retrieval: the big picture



Testing the algorithm: simulated retrievals of aerosol optical depths

O_4 SCDS at 4 wavelengths (360 nm, 477 nm, 577 nm, 630 nm)

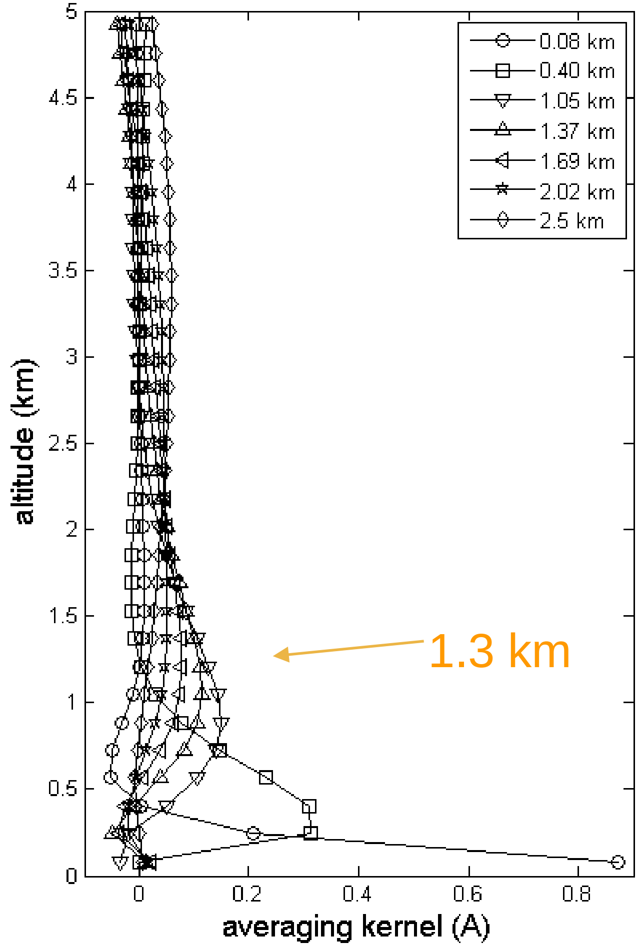
- A = O_4 DSCD
- B = O_4 DSCD +DI

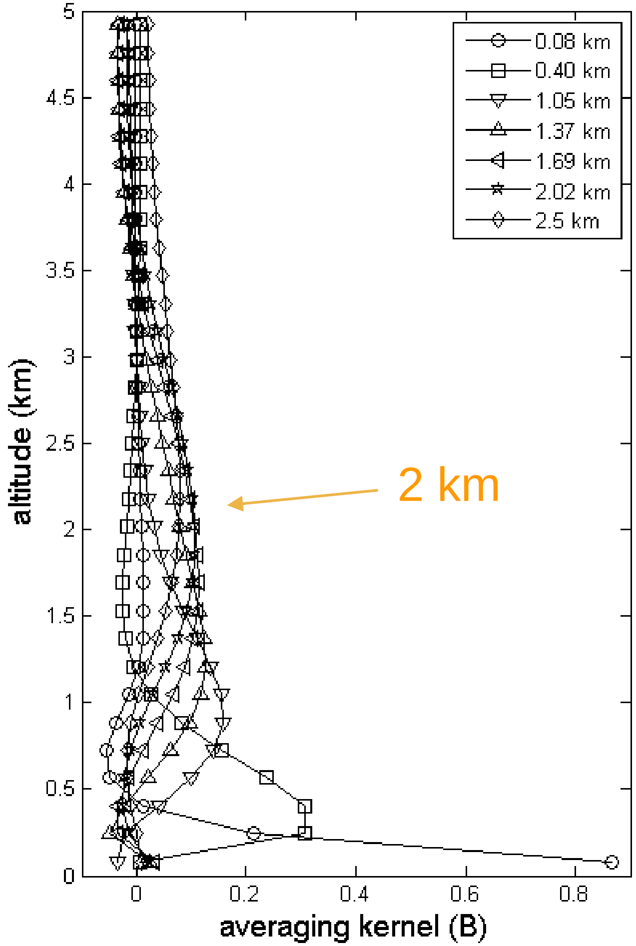


Testing the algorithm: information content for aerosol optical depth retrieval

4 wavelengths (360 nm, 477 nm, 577 nm, 630 nm)

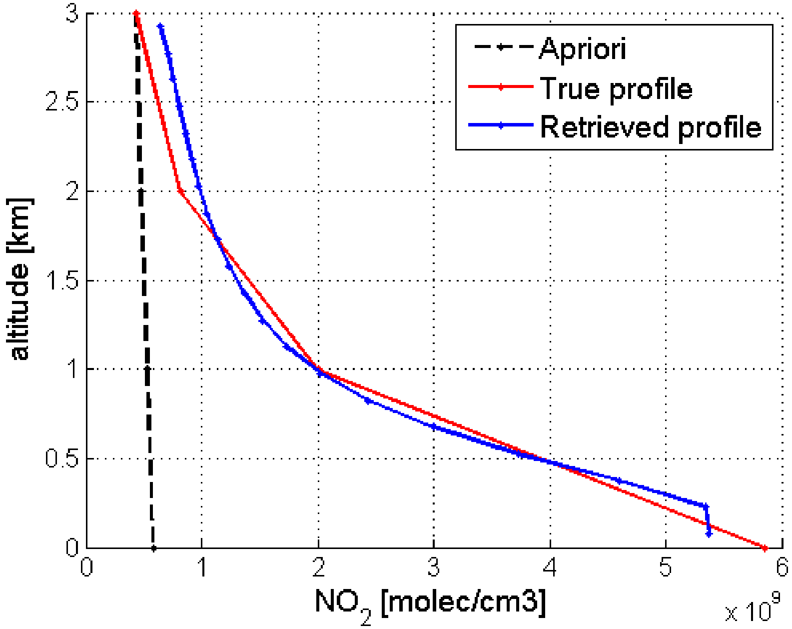
- A = O_4 DSCD
- B = O4 DSCD +DI

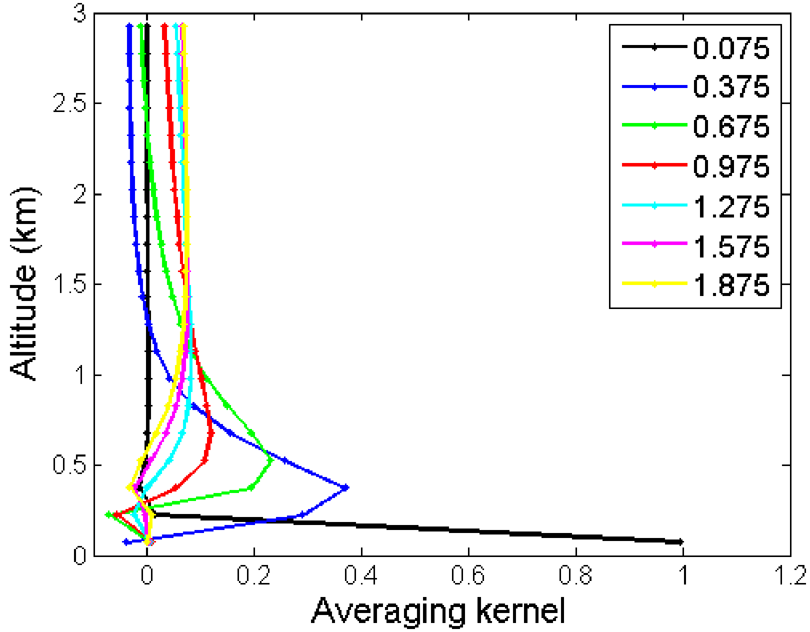




DFS = ~4

Testing the algorithm: NO_2 retrieval case





DFS = ~4

➔ Up to 4 independent pieces of information in the troposphere, depending on NO_2 profile shape and aerosol content

Measurements in Beijing (July 2008)

- Elevation angles (9):
  2°, 4°, 6°, 8°,
  10°, 12°, 15°,
  (30°), 90° (zenith)
- Pointing direction: North

DOAS settings for O_4 retrieval:

- 340-370 nm
  → O_4, O_3, NO_2, BrO, and Ring
- 455-500 nm
  → O_4, O_3, NO_2, H_2O, and Ring
- 540-588 nm
  → O_4, O_3, NO_2, H_2O, and Ring
- 602-645 nm
  → O_4, O_3, NO_2, H_2O, O_2, and Ring

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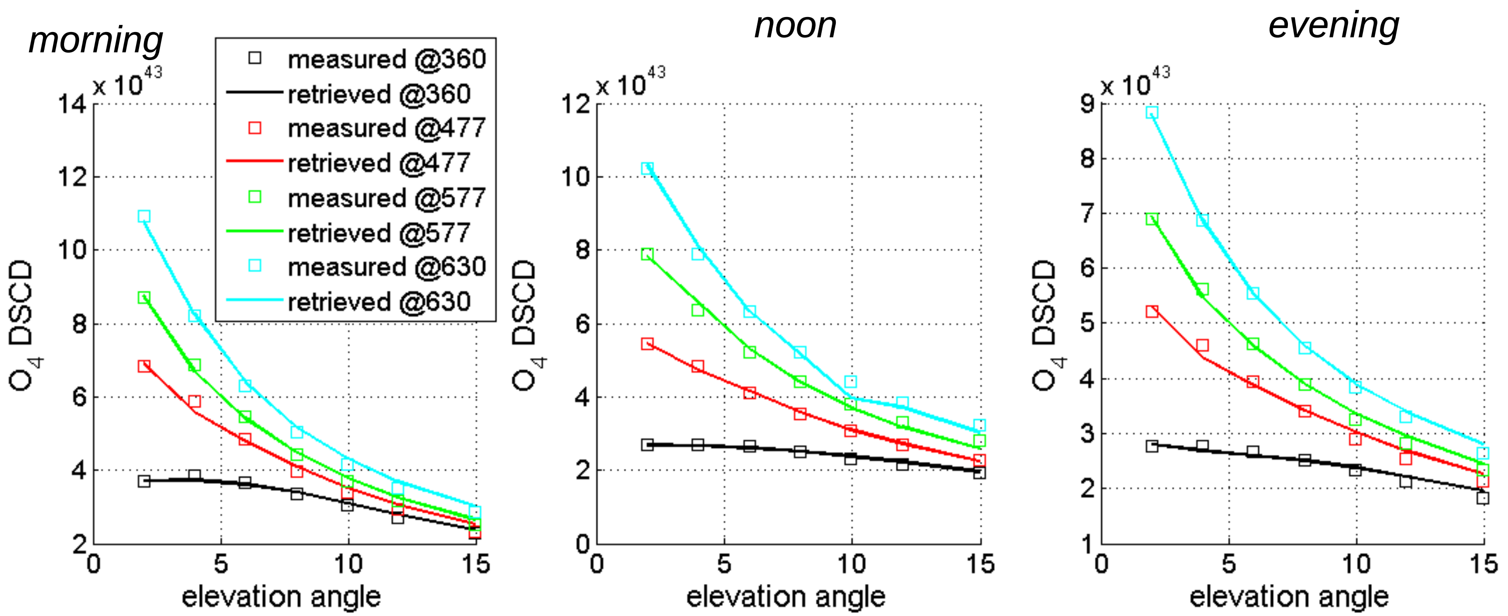
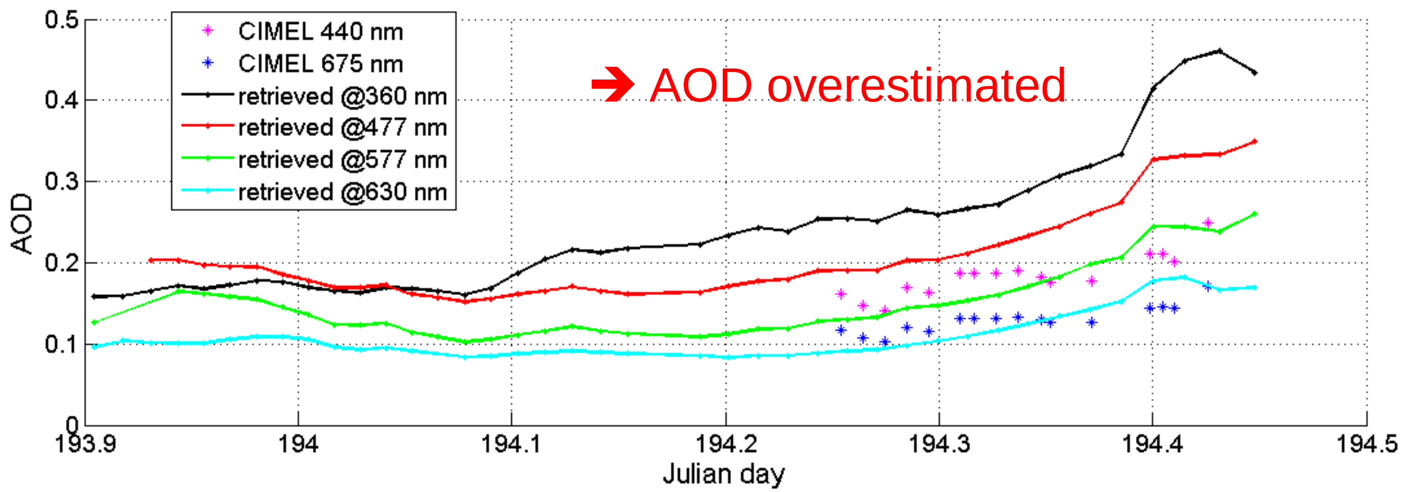
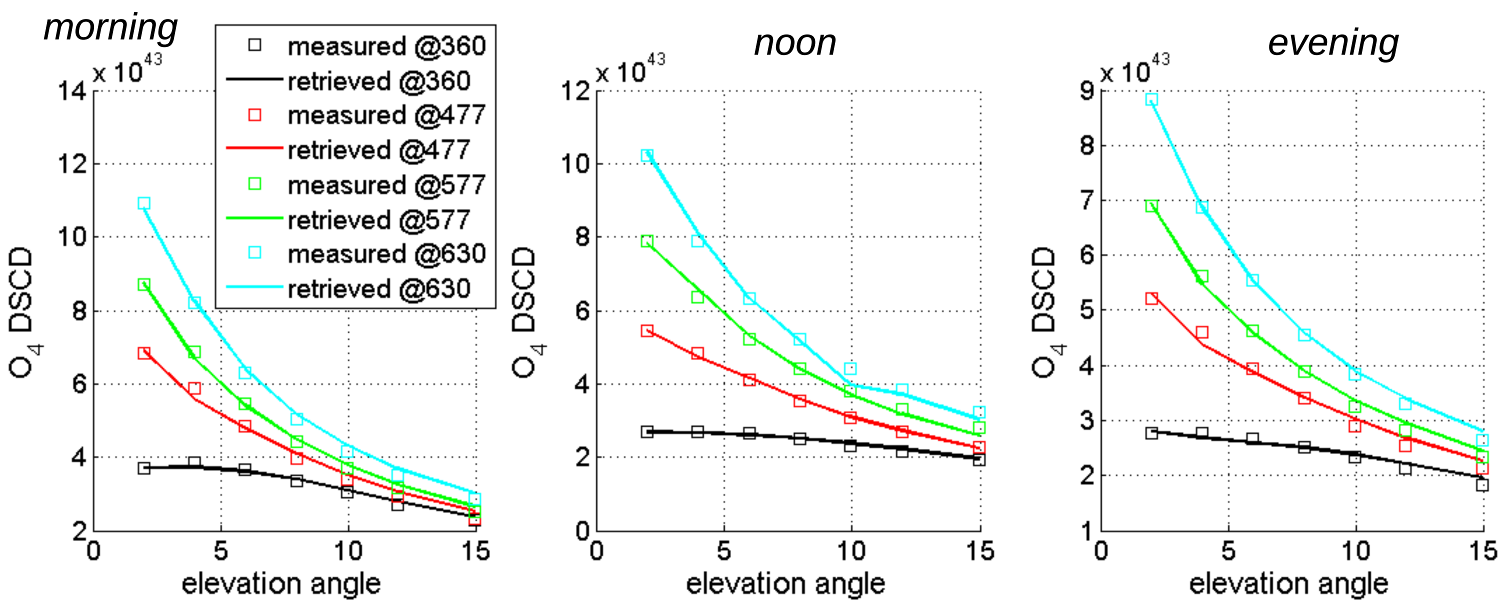
Preliminary results: AOD retrievals based on O_4 measurements





Preliminary results: Optimal Estimation



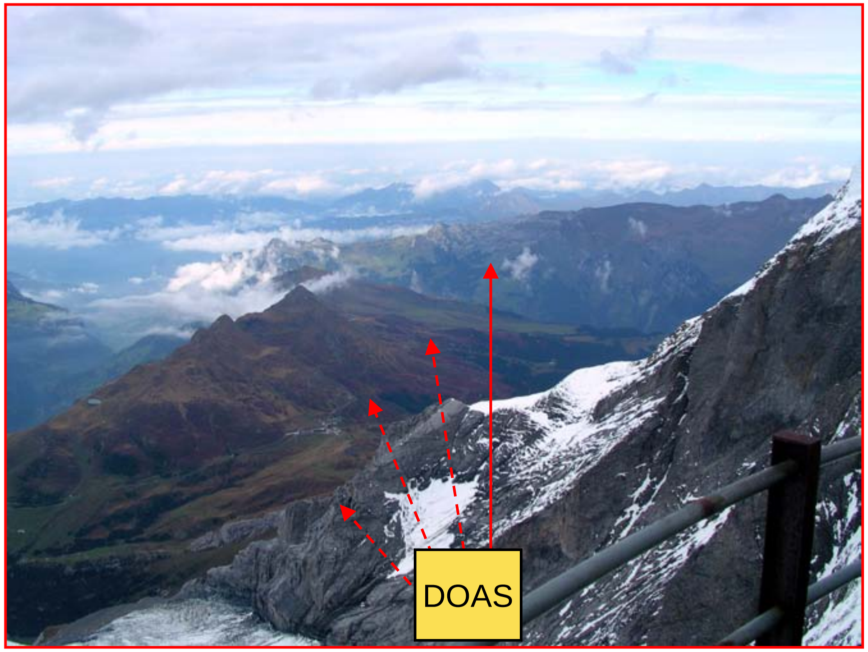
- <u>Basic problem</u>: the solution may not be unique
- Further constrain is necessary → use additional information to better constrain the AOD (intensities + direct-sun measurements)
- Add more geometries (almucantar + principal plane) to retrieve aerosol phase function

Summary and outlook (1)

- MAXDOAS instrumentation and retrieval algorithms currently under development at BIRA-IASB
- Instrument's capability demonstrated for the detection of a number of trace gases of interest for tropospheric and stratospheric chemistry (NO_2, O_3, BrO, HCHO, glyoxal, HONO, SO_2, ...)
- Straighforward exploitation possible for tropospheric column retrieval (geometrical approximation) → direct interest e.g. for satellite validation
- Information content analysis and simulated retrievals show potential for aerosol and trace gas profile retrieval in the troposphere, with up to 4 pieces of independent information on the tropospheric profile. More work needed to assess and validate the method.

Summary and outlook (2)

- Current funding available for 2 instruments in addition to Beijing one. One of them considered for installation at Jungfraujoch, in complement to SAOZ instrument
- Could complement existing in-situ instrumentation for trace gas and aerosol monitoring
- Possibility to sample down into the valley and retrieve profile information → distinguish between PBL and free troposphere?
- Possibility to improve information content by adding second MAXDOAS system in Payerne (e.g. in collaboration with EMPA)



Thank you for your attention !