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## Abstract

Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) is a widely used technique for the detection of atmospheric trace gases, e.g. NO<sub>2</sub>, SO<sub>2</sub>, but also for the oxygen collision complex O<sub>4</sub>, whose atmospheric distribution is well known. By comparing measured O<sub>4</sub> differential slant column densities (dSCDs) with modelled ones, information on aerosol distributions and optical properties can be gained<sup>1,5</sup>. In combination with a radiative transfer model<sup>4</sup>, an inversion of measured dSCDs allows the retrieval of vertical aerosol extinction profiles and properties.

## Instrumentation

### Differential Optical Absorption Spectroscopy (DOAS)

DOAS is based on the separation of narrowband differential absorption structures from broadband absorption and scattering. The differential absorption structures are characteristic for each species which allows a reconstruction of the measured spectrum to determine the concentrations of different absorbers at the same time.

$$I(\lambda) = I_0(\lambda) \cdot \exp\left(-\sum_i \sigma_i(\lambda) \cdot \bar{c}_i \cdot L - \varepsilon_p(\lambda) \cdot L\right) \rightarrow \ln(I_0(\lambda)/I(\lambda)) = \sum_i \sigma_i'(\lambda) \cdot a_i + P(\lambda)$$

$\sigma_i$ : absorption cross section of species  $i$  with concentration  $\bar{c}_i$   
 $\varepsilon_p$ : scattering (Rayleigh, Mie)  
 $L$ : measured optical depth  
 $a_i$ : Fitted model

### Multi-Axis DOAS

MAX-DOAS is a passive measurement configuration that uses scattered sunlight as light source. It measures slant column densities (SCDs) i.e. concentrations integrated along the light path. By scanning several elevation angles, vertical profiles of absorbing trace gases and aerosols for heights up to several kilometers can be retrieved using radiative transfer models and optimal estimation algorithms<sup>9-16</sup>.

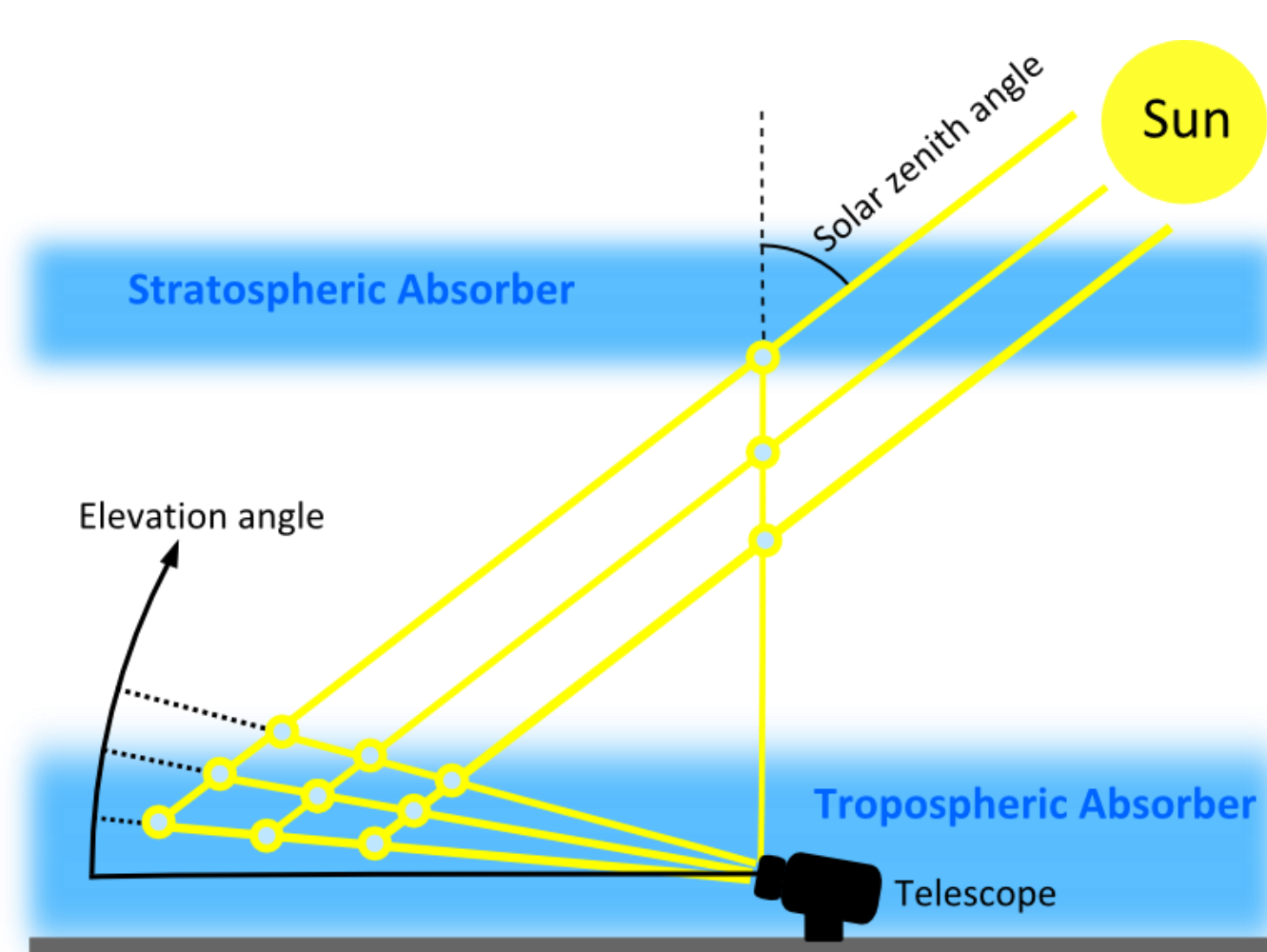
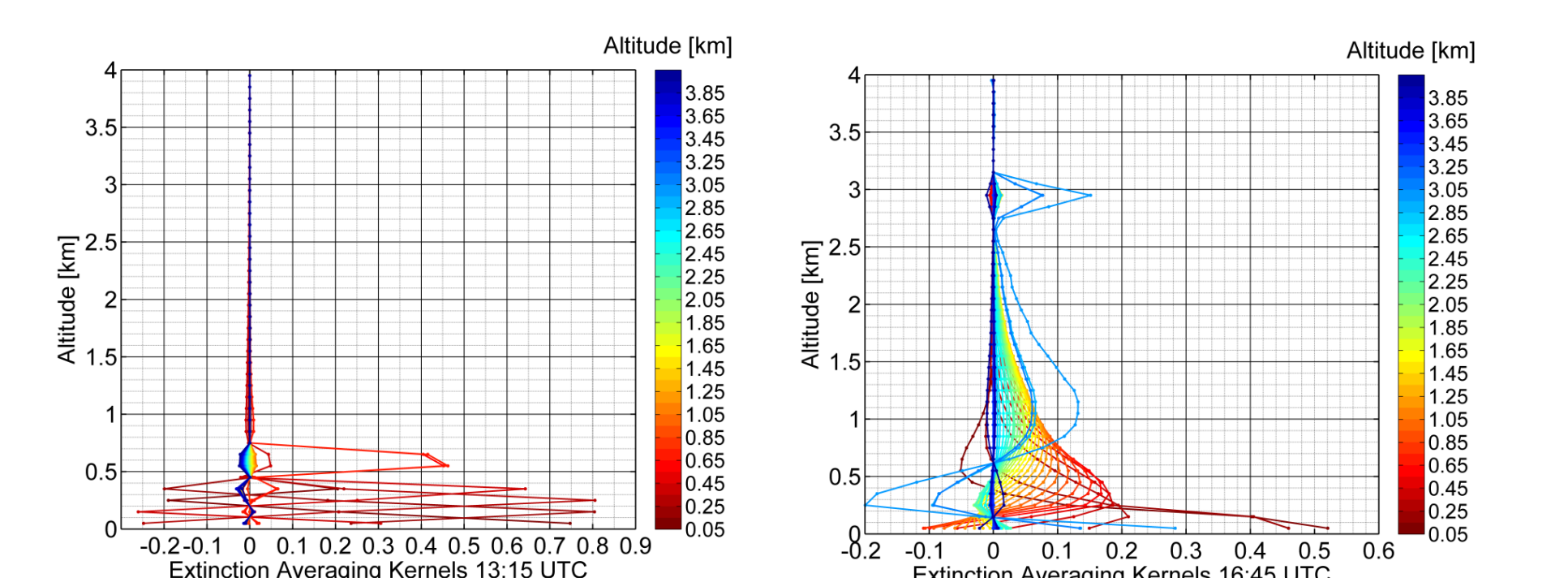
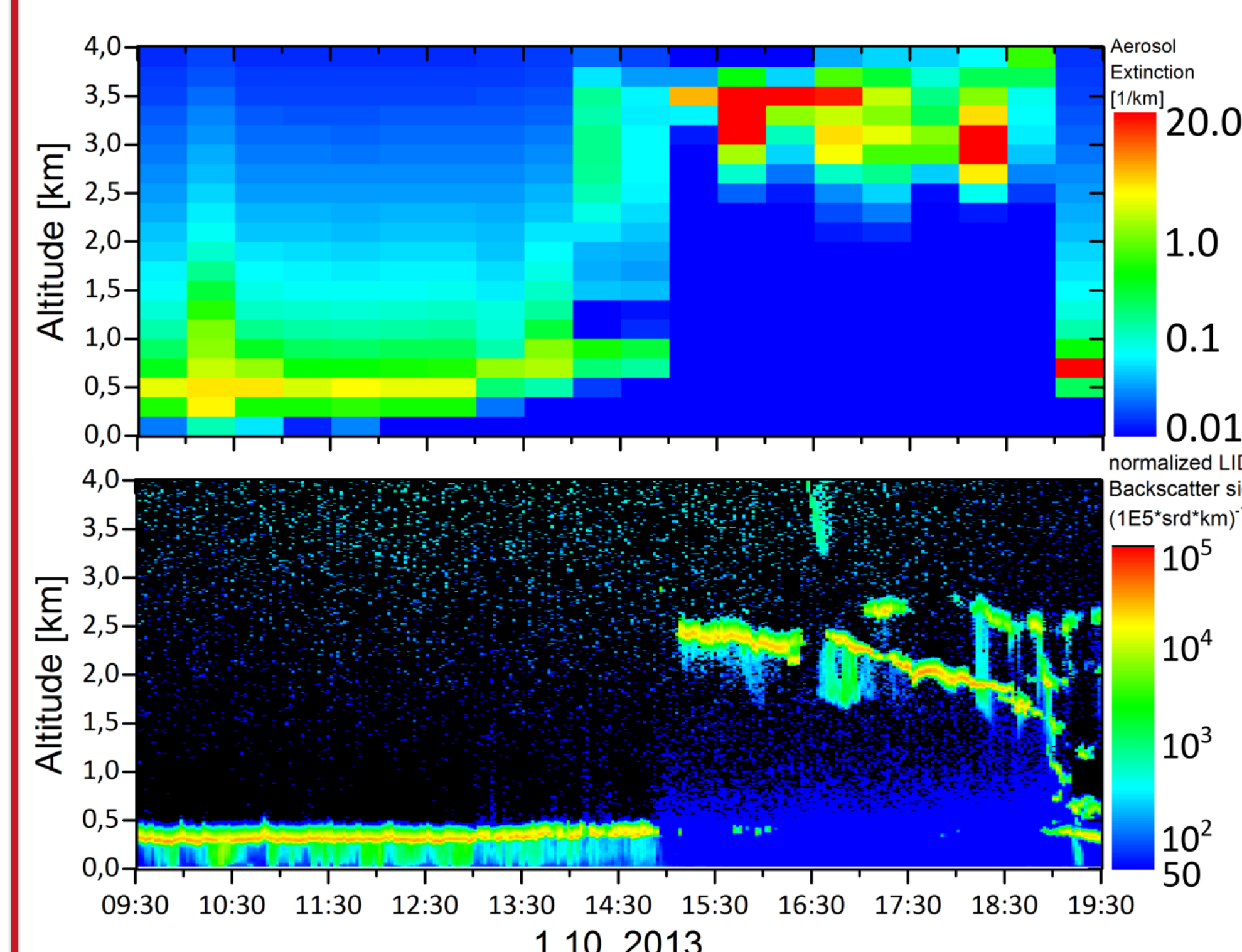


Fig.1: Principle of MAX-DOAS measurements<sup>2</sup>

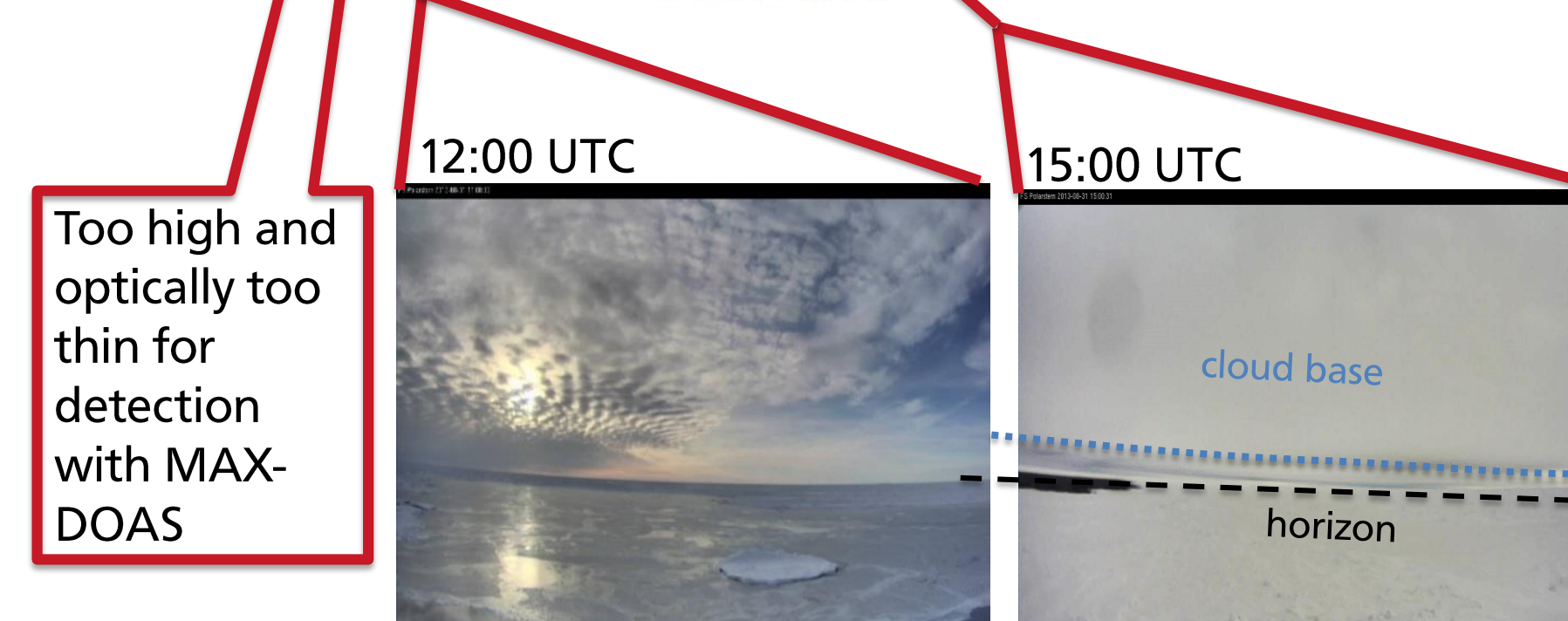
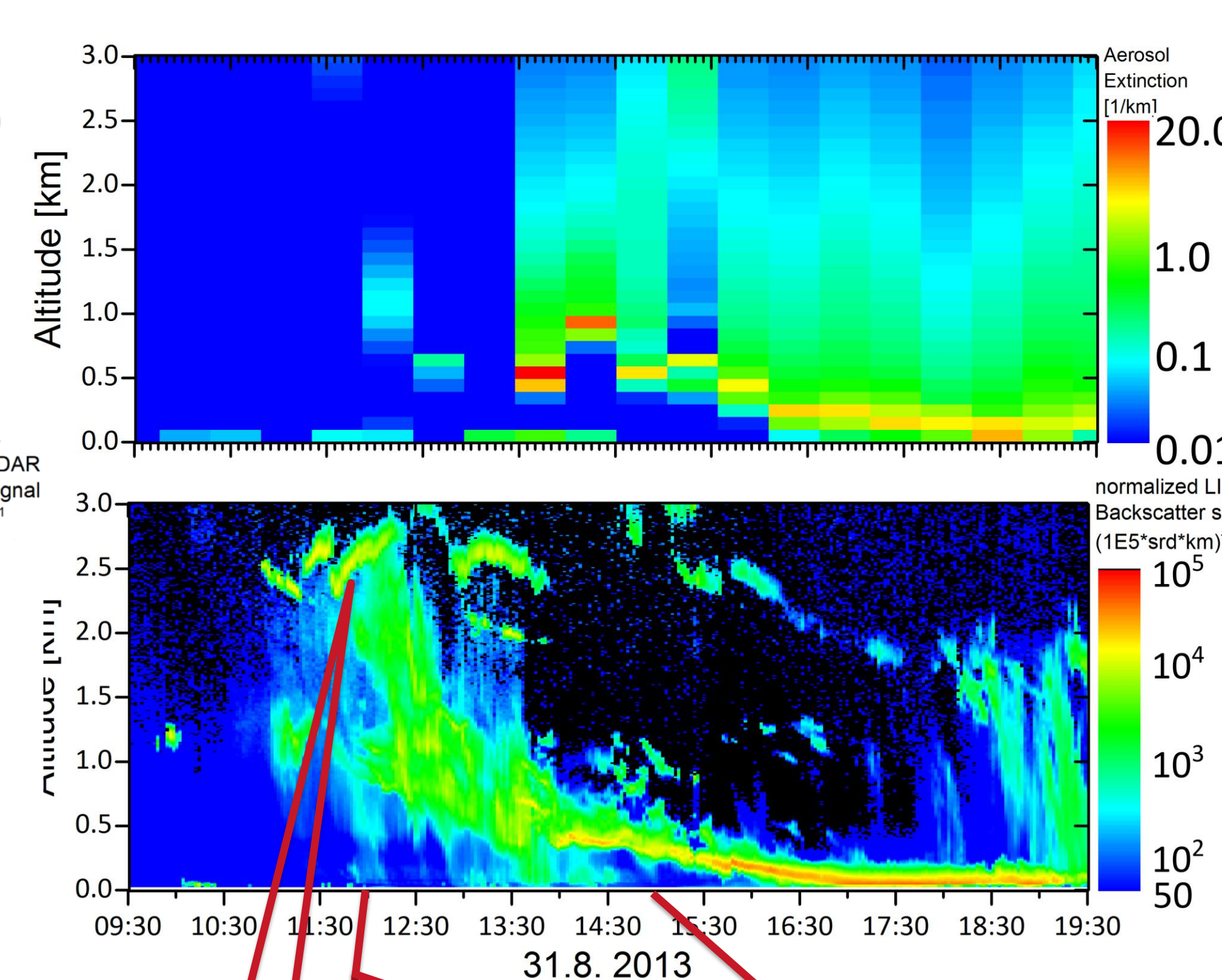
## Comparison of retrieved extinction profiles with high resolution ceilometer data

To validate the extinction profile retrieval, data of a MAX-DOAS on FS Polarstern<sup>2</sup> and a co-located VAISALA CL-51 Ceilometer<sup>3</sup> from cruise ANT XXIX-7 to the Antarctic sea ice region are used. The CL-51 is a LIDAR system operating at 910 nm. The cruise was dominated by low, optically thick clouds.

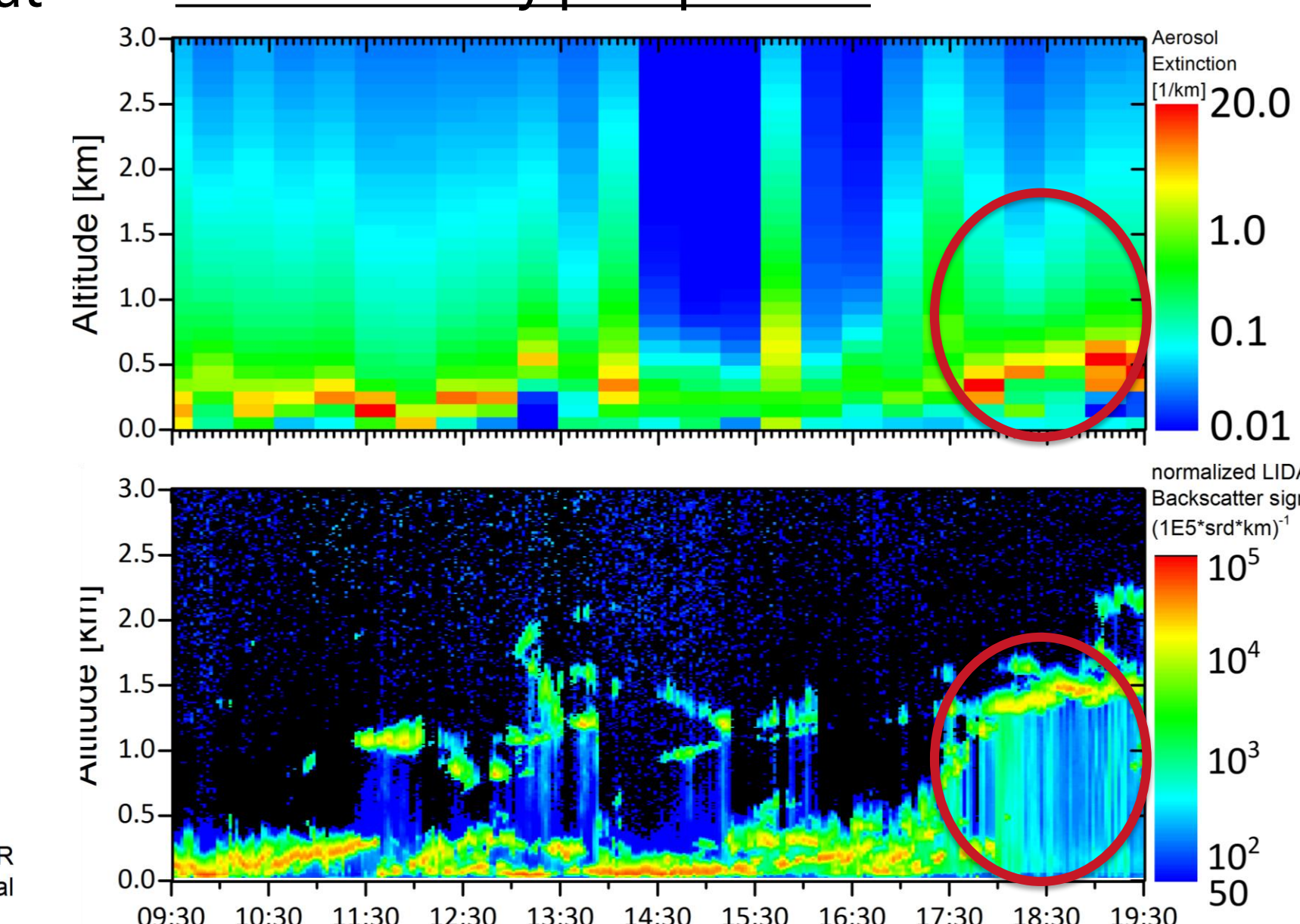
### High cloud cover vs. low cloud cover



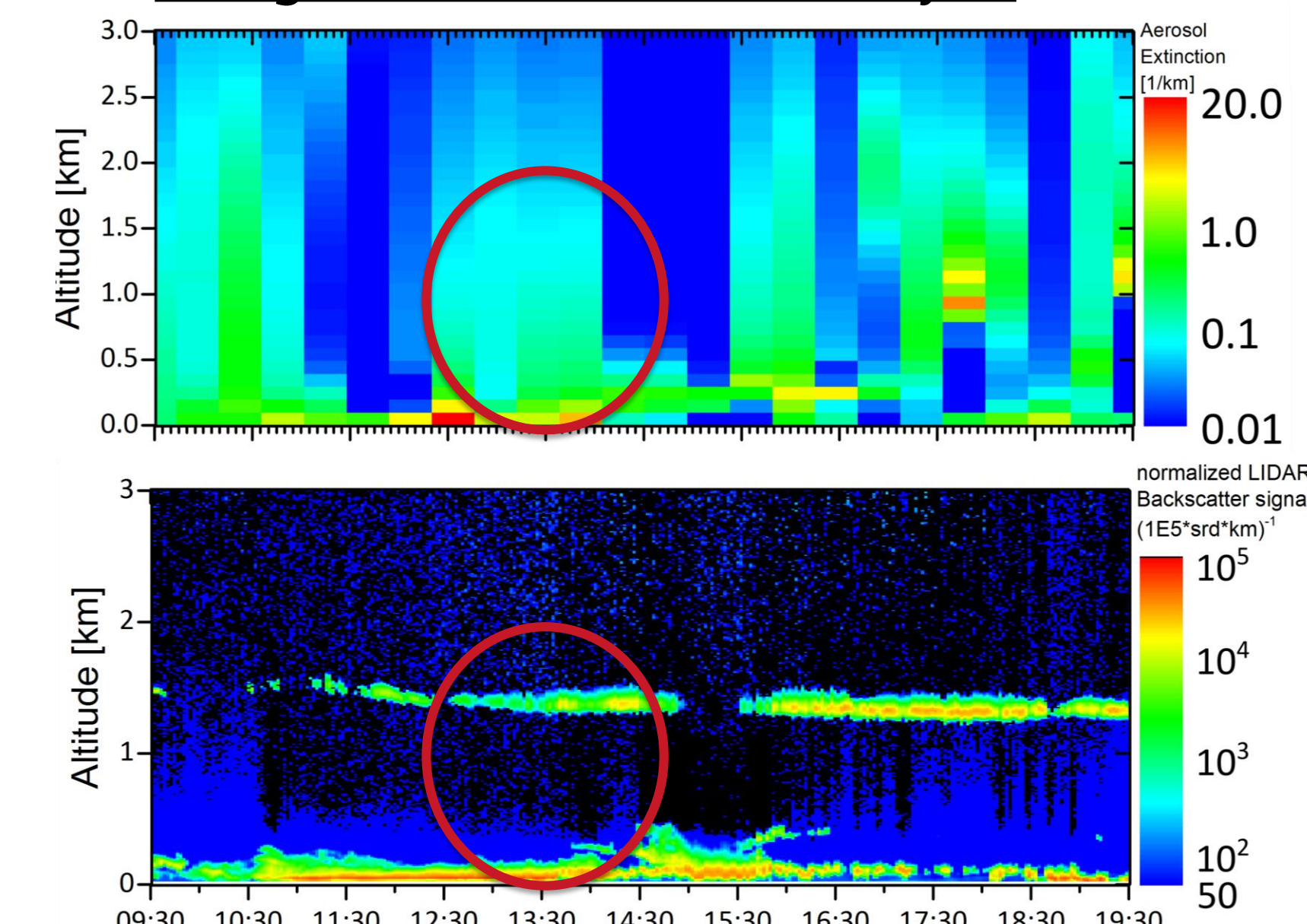
### Descending cloud cover



### Disturbance by precipitation



### No agreement for two cloud layers

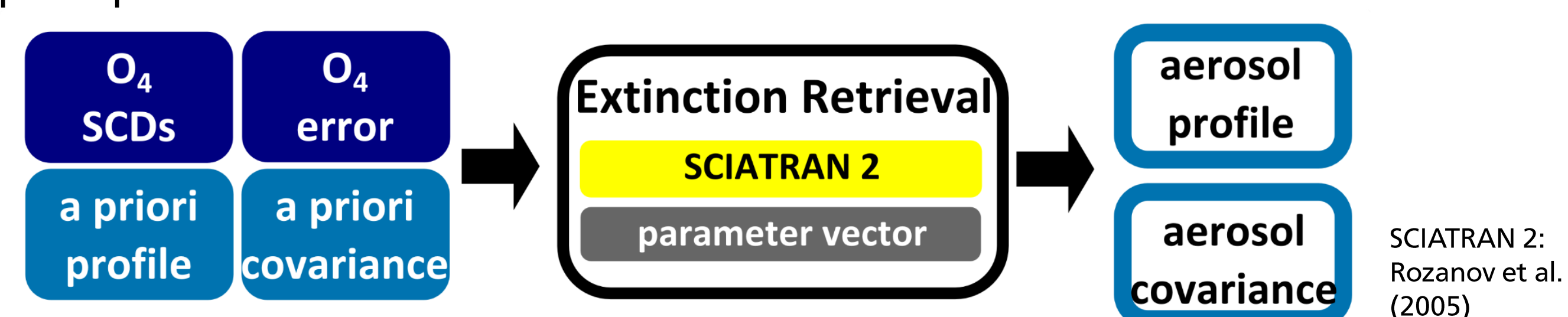


## Profile retrieval by inversion

To obtain vertical extinction profiles, the state of the atmosphere  $\vec{x}$  (i.e. the extinction profile) is inferred from the observable O<sub>4</sub> slant column densities ( $\vec{y}$ ) using the knowledge about the functional relation between both (the radiative transfer model  $F(\vec{x})$ <sup>4</sup>).

$$\vec{y} = F(\vec{x}) + \sigma_\varepsilon \rightarrow \hat{\vec{x}}$$

Since typically the information from a set of dSCDs is not sufficient to directly perform the inversion of the equation above, information from an additional a-priori profile is needed.



SCIATRAN 2:  
Roazanov et al.  
(2005)

The optimal estimation is obtained by minimizing the sum of the error-weighted differences between measured and modeled dSCDs and the actual and the a-priori profile.<sup>6</sup>

$$\chi^2(\vec{x}) = [F(\vec{x}) - \vec{y}]^T S_\varepsilon^{-1} [F(\vec{x}) - \vec{y}] + [\vec{x} - \vec{x}_a]^T S_a^{-1} [\vec{x} - \vec{x}_a]$$

The retrieved state of the atmosphere therefore contains information from both measurement and a-priori estimate. The relation of both contributions in the retrieval  $\hat{\vec{x}}$  can be visualized with the averaging kernel matrix  $\hat{A}$ :

$$\hat{\vec{x}} = \hat{A}\vec{x} + (I_n - \hat{A})\vec{x}_a + G\sigma_\varepsilon$$

The lines of the Averaging Kernel matrix are curves that describe from which altitudes extinction contributes to the retrieved profile in a certain altitude as well as the relative contribution of information by the measurement. Ideally the kernels peak in their respective altitude and are zero everywhere else.

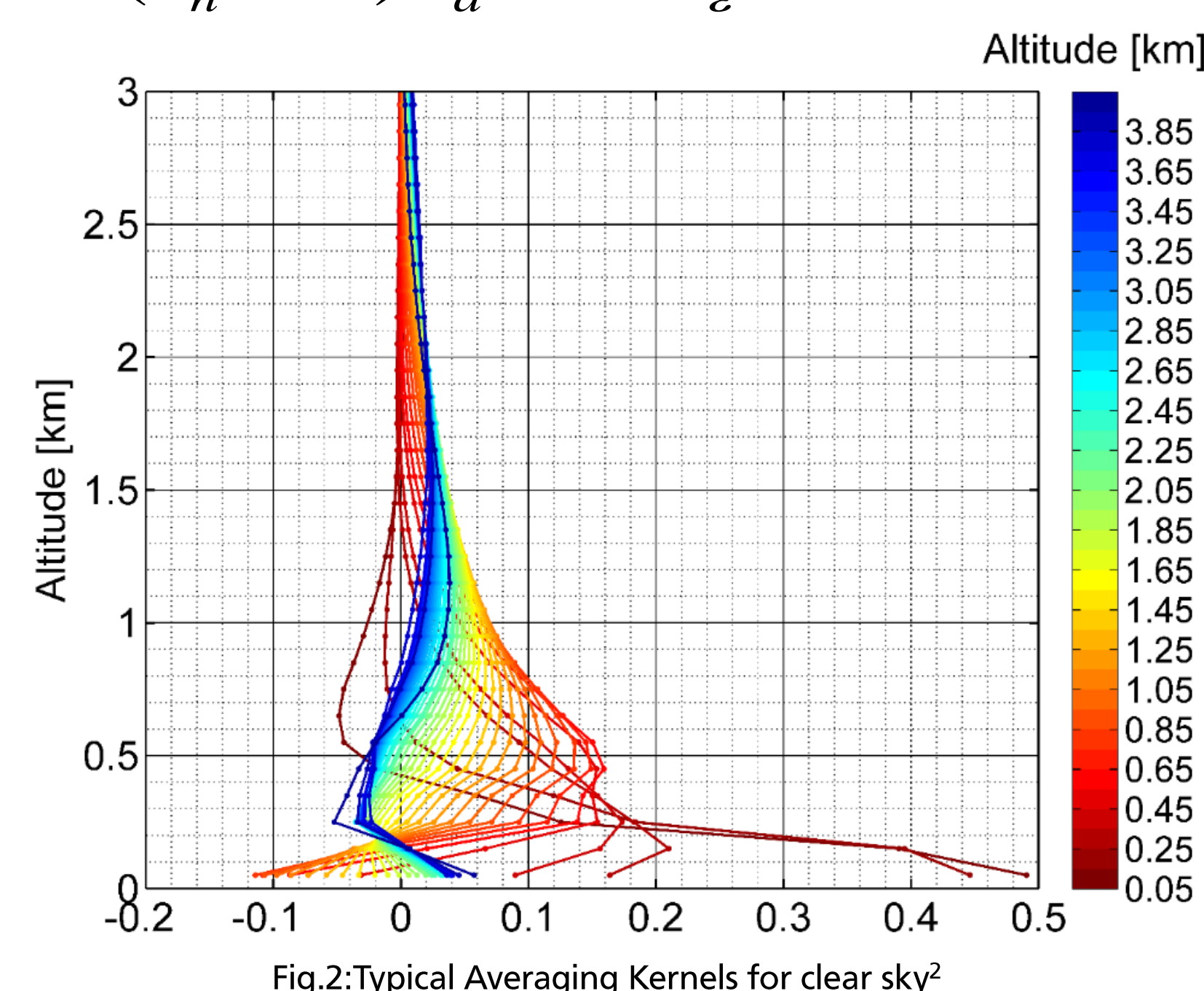


Fig.2: Typical Averaging Kernels for clear sky<sup>2</sup>

## Conclusions

- MAX-DOAS O<sub>4</sub> measurements can accurately detect the cloud altitude up to 1.5km altitude. Cloud optical thickness yields realistic results but could not be validated.
- For optically thick and homogeneous cloud covers, the vertical sensibility is even extended to 2km and more because of non-linear influence of extinction on O<sub>4</sub> slant column densities
- Precipitation and multiple cloud layers so far lead to erroneous extinction profiles
- For homogeneous cloud cover, a trace gas retrieval using the retrieved extinction profiles is possible.
- Below homogeneous clouds, ground-based MAX-DOAS measurements are very sensitive and have a higher vertical resolution than under clear sky conditions.

## References

1. Friß, U., P. S. Monks, J. J. Remedios, A. Rozanov, R. Sinreich, T. Wagner, and U. Platt (2006). "MAX-DOAS O<sub>4</sub> measurements: A new technique to derive information on atmospheric aerosols: 2. Modeling studies". In: Journal of Geophysical Research 111.D14, p. D14203.
2. Nasse, J.-M. (2014). "Retrieval of aerosol and trace gas vertical profiles in the Antarctic troposphere using helicopter- and ship-borne MAX-DOAS measurements". Master thesis, Heidelberg University
3. König-Langlo, G. (2014). Ceilometer CL51 raw data measured during POLARSTERN cruise ANT-XXIX/7, links to raw data files. Dataset #833802 (DOI registration in progress). Tech. rep. Bremerhaven: Alfred Wegener Institute, Helmholtz Center for Polar and Marine Research.
4. Roazanov, A., V. Roazanov, M. Buchwitz, A. Kokhanovsky, and J.P. Burrows (2005). "SCIATRAN 2.0 - A new radiative transfer model for geophysical applications in the 175-2400nm spectral region". In: Advances in Space Research 36.5, pp. 1015-1019
5. Wagner, T., B. Dix, C. v. Friedeburg, U. Friß, S. Sanghavi, R. Sinreich, and U. Platt (2004). "MAX-DOAS O<sub>4</sub> measurements: A new technique to derive information on atmospheric aerosols—Principles and information content". In: Journal of Geophysical Research 109.D22205
6. Yilmaz, S. (2012). "Retrieval of Atmospheric Aerosol and Trace Gas Vertical Profiles using Multi-Axis Differential Optical Absorption Spectroscopy". Dissertation, Heidelberg University.