Retrieval of Aerosol Profiles using Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS)

Selami Yilmaz[1], Udo Frieß[1], Arnoud Apituley[2], Gerrit de Leeuw[3,4,5], Bas Henzing[5] and Ulrich Platt[1]

[1]*Institute of Environmental Physics, University of Heidelberg, Germany, selami.yilmaz@iup.uni-heidelberg.de*
[2]*RIVM – National Institute for Public Health and the Environment, Bilthoven, The Netherlands, arnoud.apituley@rivm.nl*
[3]*Finnish Meteorological Institute, Helsinki, Finland, gerrit.leeuw@fmi.fi*
[4]*Department of Physics, University of Helsinki, Finland*
[5]*TNO, Utrecht, The Netherlands, bas.henzing@tno.nl*

ABSTRACT

Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) allows to derive substantial information on the aerosol extinction profile and optical/microphysical properties by passive remote sensing. While established passive remote sensing techniques using sun photometers only provide the total optical depth, information on the aerosol vertical distribution is derived from measurements of the optical depth of the oxygen dimer (O_4) in addition to the (relative) intensity of diffuse skylight at different viewing directions and wavelengths by MAX-DOAS.
Here we present results from MAX-DOAS aerosol measurements during the EUCAARI IMPACT campaign in Cabauw, Netherlands in May 2008. The comparison to established techniques measuring aerosol optical properties like the extinction profile and the total optical depth demonstrates that MAX-DOAS has the capability to derive information on atmospheric aerosols and represents a powerful measurement technique. Here we present the idea of this new technique and results of the intercomparison.

1 INTRODUCTION

Aerosols are enormously influencing chemical and physical processes in the Earth's atmosphere. The latest IPCC report shows that aerosols have a significant impact on the atmospheric energy budget, inter alia by modifying the amount and optical properties of clouds. But the uncertainties in the assumptions and the comparatively low level of scientific understanding demand more intensive efforts in terms of collecting atmospheric aerosol properties, as well as new measurement techniques providing advantages like cost efficiency, simple and robust instrumentation, enabling the integration in worldwide measurement networks.

DOAS (Differential Optical Absorption Spectroscopy) is a well established technique for the measurement of trace gas concentrations in the atmosphere [1]. Multi Axis DOAS is a relatively new technique which was developed in recent years [2] and enables the determination of the vertical distribution of several UV and vis light absorbing trace gases like NO_2, SO_2, BrO, HCHO and glyoxal. This potential was demonstrated successfully by several groups. By measuring trace gases with a known vertical distribution like the oxygen dimer O_4 and subsequently using inverse modelling methods, it is possible to retrieve atmospheric aerosol profiles [3,4].

2 MAX-DOAS

The idea of the passive MAX-DOAS technique is to collect scattered sunlight from different viewing directions. This is described by the Lambert-Beer law:

$$I(\lambda, L) = I_0(\lambda) \cdot e^{-\int_0^L \left[\sum_j \sigma_j(\lambda) \cdot c_j(l) + \varepsilon_M(\lambda, l) + \varepsilon_R(\lambda, l) \right] dl} \tag{1}$$

where I_0 is the unattenuated intensity, σ_j the absorption cross section and c_j the concentration of trace gas j. ε_M and ε_R are the extinction coefficient for Rayleigh and Mie scattering, respectively. The basic measured quantity is the slant column density S_j, the integrated concentration along the light path,

$$S_j(L) = \int_0^L c_j(l) dl \tag{2}$$

or the respective optical depth τ_j

$$\tau_j(\lambda, L) = \sigma_j(\lambda) \cdot S_j(L) \tag{3}$$

In the analysis procedure the linear least squares fit and the non-linear Levenberg-Marquardt method is used to fit scaled absorption cross sections of several trace gases with absorption structures, in the relevant wavelength range to the measured spectra.

$$\chi^2 = \int_{\lambda_1}^{\lambda_2} \left[\ln\left(I(\lambda, L)\right) - \ln\left(I_0(\lambda)\right) + \sum_j \sigma_j(\lambda) \cdot S_j(L) + \sum_m p_m \cdot \lambda^m \right]^2 d\lambda \xrightarrow{!} \min \tag{4}$$

where the last term accounts for broad band absorptions from trace gases and for Rayleigh and Mie scattering. Measurements in zenith direction are used as the reference spectra I_0, therefore the determined quantities are differential slant column densities.

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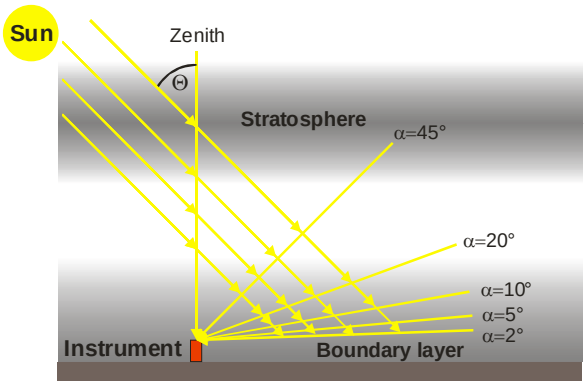

Figure 1. Simplified sketch of the viewing geometry of a MAX-DOAS measurement

During a MAX-DOAS measurement, the telescope is pointing to several directions with different elevation angles (Figure 1). Thus, the vertical distribution of the trace gases in the lower troposphere can be retrieved. Moreover, the light paths at low elevation angles are relative long in the lowermost atmospheric layer, leading to high sensitivity to trace gases in this layer.

3 INVERSE MODELLING

In contrast to forward modelling procedures, where a radiative transfer model is used to simulate the measurement, the inverse modelling approach is used to estimate the atmospheric state of interest in that lead to the performed measurement results. In practise, this means the minimization of the cost function

$$\chi^2 = \left[\mathbf{F}(\mathbf{x})-\mathbf{y}\right]^T \mathbf{S}_\varepsilon^{-1}\left[\mathbf{F}(\mathbf{x})-\mathbf{y}\right]+ \\ +\left[\mathbf{x}-\mathbf{x}_a\right]^T \mathbf{S}_a^{-1}\left[\mathbf{x}-\mathbf{x}_a\right] \xrightarrow{!} \min \quad (5)$$

with the measurement vector $\mathbf{y}$ containing in our case the O_4 optical depth and the relative changes in skylight intensity as a function of wavelength and viewing geometry. $\mathbf{F}$ is the forward model, in our case the radiative transfer model Sciatran [5]. $\mathbf{x_a}$ is the a priori state vector and $\mathbf{x}$ is the best estimation for the atmospheric state vector, consisting of the aerosol extinction profile and microphysical properties. $\mathbf{S_e}$ and $\mathbf{S_a}$ are the covariance matrices containing the errors of the measurement and the a priori state, respectively. The solution is determined iteratively using the Gauss-Newton method

$$\mathbf{x}_{i+1} = \mathbf{x}_i + \left(\mathbf{S}_a^{-1} + \mathbf{K}_i^T \mathbf{S}_\varepsilon^{-1} \mathbf{K}_i\right)^{-1} \cdot \\ \cdot \left[\mathbf{K}_i^T \mathbf{S}_\varepsilon^{-1}\left(\mathbf{y}-\mathbf{F}(\mathbf{x}_i)\right) - \mathbf{S}_a^{-1}\left(\mathbf{x}_i-\mathbf{x}_a\right)\right] \quad (6)$$

with the weighting function

$$K_{jk} = \frac{\partial F_j(\mathbf{x})}{\partial x_k} \quad (7)$$

The retrieval has been performed by simultaneously using O_4 differential slant column densities and relative intensities at three wavelengths (360, 477 and 577 nm), observed at elevation angles of 90°, 20°, 10°, 5° and 2°. Retrievals have been performed in time steps of 15 minutes. Retrieved quantities are:

- aerosol extinction profiles at a reference wavelength of 550 nm, on a vertical grid of 200 m in the lowermost 4000 m of the atmosphere
- the composition of the particles, parameterised by a mixture of reflecting sulphate aerosols and absorbing black carbon.

4 CAMPAIGN AND INSTRUMENTATION

Our newly developed, cost effective MAX-DOAS instrument consists of the telescope unit which is connected via a quarz fiber bundle to the separate spectrometer unit, making it possible to operate indoors under stable conditions (Figure 2). The usage of two rotatable quartz prisms enables the possibility to collect light from any direction. A spherical mirror focuses the light without spectral aberration, which is necessary, since the three temperature stabilized miniature spectrographs cover the wide wavelength range of 290 -790 nm. A built-in embedded PC supports the fully automated operation and remote control via TCP/IP.

The intensive observation period of the EUCAARI IMPACT campaign in Cabauw – The Netherlands took place during the whole May 2008. The main focus of this campaign was the measurement of several different chemical and physical properties of atmospheric aerosols. Amongst all the different instruments, also ground based raman lidar and sun photometer measurements were performed simultaneously to our MAX-DOAS measurements, enabling the comparison of the established to our new technique as part of the joint research activity in the scope of the EUSAAR project.

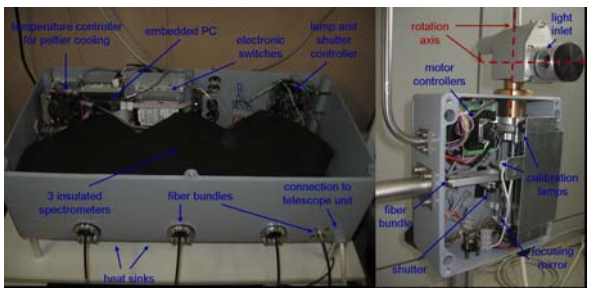

Figure 2. *MAX-DOAS instrument consisting of a spectrometer (left) and telescope (right) unit.*

5 RESULTS AND DISCUSSION

5.1 Qualitative Comparison to lidar data

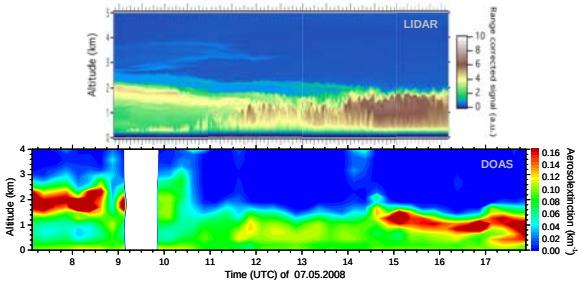

Figure 3. *Comparison of Lidar signal (top) with MAX-DOAS aerosol extinction profiles on May 7, 2008.*

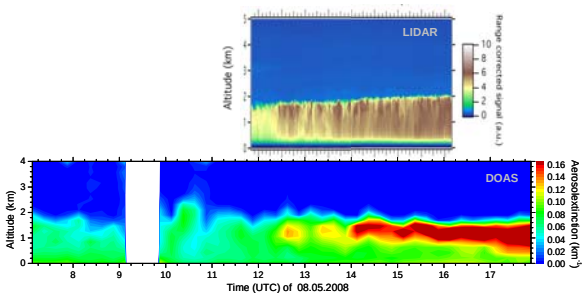

Figure 4. *Comparison of Lidar signal (top) with MAX-DOAS aerosol extinction profiles on May 8, 2008*

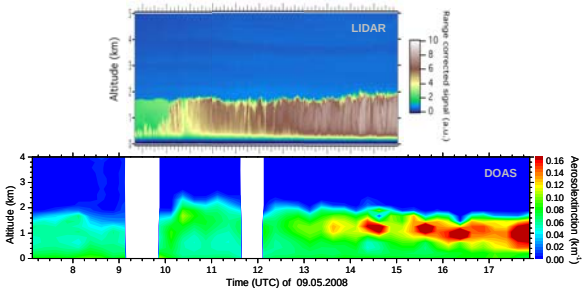

Figure 5. *Comparison of Lidar signal (top) with MAX-DOAS aerosol extinction profiles on May 9, 2008.*

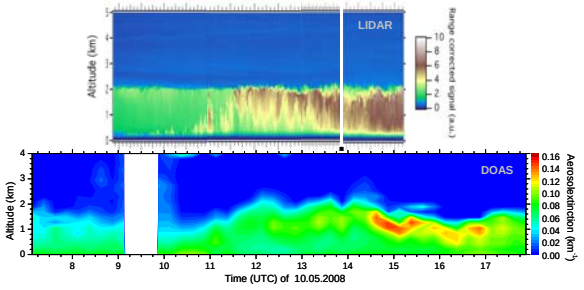

Figure 6. *Comparison of Lidar signal (top) with MAX-DOAS aerosol extinction profiles on May 10, 2008.*

Figures 3 - 6 show examples for the comparison of the aerosol extinction profiles retrieved from MAX-DOAS with the range corrected signal from the Raman Lidar Ceali of RIVM in Cabauw. The general structure of the boundary layer, i.e. the boundary layer height, the increase in extinction in the afternoon, and features such as an elevated aerosol layer in the morning of May 7, are well captured by the MAX-DOAS measurements. This demonstrates the capability of MAX-DOAS measurements to determine the vertical structure of the planetary boundary layer.

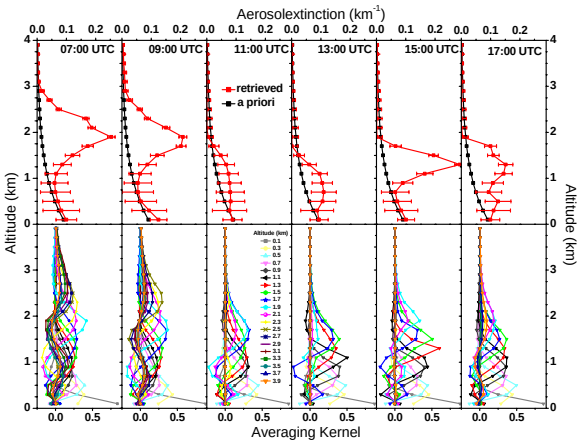

Figure 7. *Individual aerosol extinction profiles from MAX-DOAS (top) and corresponding averaging kernels (bottom) on May 7, 2008.*

The vertical resolution of the MAX-DOAS measurements is quantified by the averaging kernels shown in Figure 7. The vertical resolution decreases with altitude, with 200 m at the surface and approximately 700 m at 1 km altitude. Note that, owing to the non-linearity of the inversion, the vertical resolution strongly depends on the vertical distribution of aerosols, with highest sensitivity at altitudes where high extinction is present.

5.2 Quantitative Comparison to Sun photometer data

The total aerosol optical thickness from MAX-DOAS has been determined by integrating the retrieved extinction profiles. Stratospheric aerosol has been adapted from a climatology based on SAGE II data, and a typical background extinction has been assumed for the free troposphere.

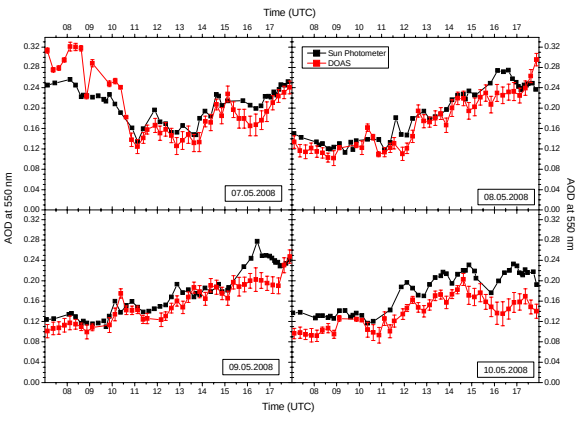

Figure 8. *Comparison of aerosol optical thickness from MAX-DOAS (red) and AERONET Sun photometer (black).*

The AOT from MAX-DOAS is in good agreement with Aeronet sun photometer measurements (Figure 8), except for the morning of May 7, when an elevated aerosol layer was present (see Figure 3) and MAX-

DOAS values are higher than AOTs from Sun photometer, and on May 10, when AOTs from MAX-DOAS are generally lower than from Sun photometer. Overall, AOT from MAX-DOAS is about 15% smaller than from the Aeronet Sun photometer, as illustrated in Figure 9.

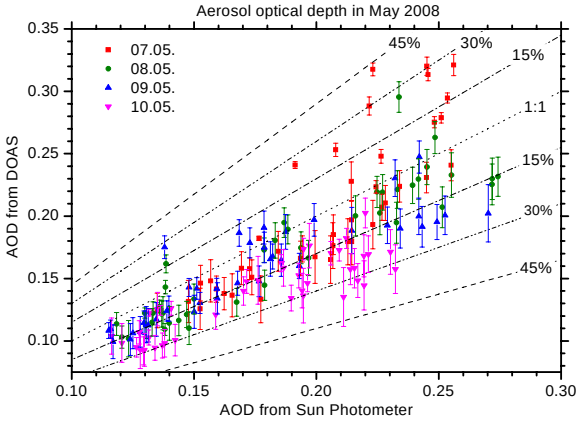

Figure 9. *Overall comparison of AOT from MAX-DOAS and AERONET Sun photometer*

6 CONCLUSION

In summary, the comparison to lidar and Sun photometer measurements show that MAX-DOAS is a valuable tool for the retrieval of atmospheric aerosol properties. The vertical structure of the boundary layer is captured well, and the retrieved aerosol optical thickness agrees well with measurements from Sun photometer. The amount of aerosols was relatively low during the EUCAARI IOP campaign. These are conditions under which the information content of MAX-DOAS measurements with respect to aerosols is relatively low, and better performance is expected for higher aerosol amounts [4]. The quantitative comparison to aerosol extinction profiles from Raman lidar is still ongoing and detailed sensitivity studies will show to which extent retrieval parameters, systematic and measurement errors have influences on the retrieved quantities. MAX-DOAS systems have the capability of fully automated operation and the necessary hardware is mobile, robust and cost-effective. With these properties, MAX-DOAS instruments are well suited to be integrated in worldwide monitoring networks.

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