MAX-DOAS O_4 measurements – a new technique to derive information on atmospheric aerosols

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Abstract. Multi AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) observations of the oxygen dimer O_4 are presented which can serve as a new method for the determination of atmospheric aerosol properties. Like established methods e.g. sun radiometer and LIDAR measurements MAX-DOAS O_4 observations determine optical properties of aerosol under atmospheric conditions (not dried). However, the novel technique has two major advantages: It utilizes the differential O_4 absorption structures and thus does not require absolute radiometric calibration. In addition, MAX-DOAS O_4 observations provide a new kind of information: since the atmospheric O_4 profile depends strongly on altitude, they can yield information on the atmospheric light path distribution and in particular on the atmospheric aerosol profile. We found that MAX-DOAS O_4 observations are a very sensitive method: even aerosol extinction below 0.001 can be detected.

Introduction

Atmospheric aerosols are important for various aspects of atmospheric chemistry and physics, they have in particular strong effects on the atmospheric radiation budget. Thus, human induced changes of the atmospheric aerosol load and its composition significantly contributes to climate change. The lack of detailed knowledge on the optical properties of aerosols causes one of the largest uncertainties in climate forcing assessments [*Houghton et al.,* 2001]. Existing methods include in-situ instruments like optical particle counters and remote sensing methods like sun photometers and Light detection and ranging (LIDAR) instruments.

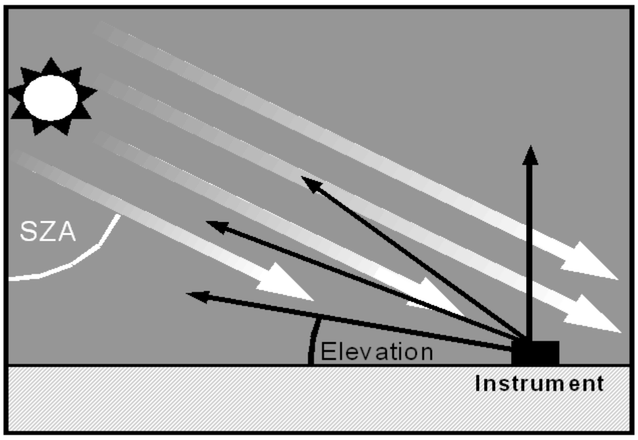

Figure 1. MAX-DOAS observation geometry. Scattered sunlight is observed under various elevation (and possibly also azimuth) angles. Often the elevation angle of the telescopes is defined with respect to the ground. The solar zenith angle (SZA) is defined as the angle between the incident sunlight and the zenith.

Here we present a new method to derive aerosol information from passive optical observations using the DOAS method [*Platt*, 1994]. We show that from this method even information on the aerosol profile can be retrieved. This method was already applied to zenith sky observations at various wavelengths [*Wagner et al*., 2002]. Here we extend the method to Multi-Axis-DOAS (MAX-DOAS) observations [*Hönninger and Platt*, 2002; *Leser et al.,* 2003; *Bobrowski et al*., 2003; *van Roozendael et al.,* 2003; *Wittrock et al.,* 2003; *Hönninger et al.,* 2004; *Heckel et al.,* 2004], which observe scattered sun light from a variety of viewing directions (see Fig. 1).

Analyzing the differential O_4 absorption from MAX-DOAS spectra has several advantages:

a) Since the MAX-DOAS O_4 analysis is a differential method, no absolute calibration is needed. In particular, degradation effects do not significantly influence the observed differential absorptions.

b) The observed O_4 absorption allows to conclude on the atmospheric distribution of photon paths, since the atmospheric O_4 concentration profile is well known and nearly constant. According to their altitude distribution aerosols change the atmospheric photon path distribution in a characteristic way and thus from MAX-DOAS O_4 observations information on the atmospheric aerosol profile can be derived.

c) O_4 absorption bands are distributed over the entire UV and visible wavelength range [*Greenblatt et al*., 1990]. Thus it is possible to derive information on the wavelength dependence of various aerosol effects.

MAX-DOAS observations during a period of increasing aerosol load

The effect of aerosols on MAX-DOAS O_4 observations can be well studied during a period of four mostly clear days during the FORMAT II campaign in Milan, Italy in September, 2003 (see http://www.nilu.no/format) when the aerosol load was strongly increasing. At the beginning the atmosphere was very clear (even the Monte Rosa mountain at a distance of about 120 km was clearly visible). On the last day even the southern edge of the Alpine mountains at a distance of only about 30 km was not visible anymore. The magnitude of the MAX-DOAS O_4 absorption and also the amplitude of the diurnal variation change significantly during these days (Fig. 2). These modifications are very sensitive to the aerosol optical depth and also on the vertical aerosol profile which was also confirmed by radiative transport modeling [*Wagner et al.,* 2004]. We also found that from MAX-DOAS O_4 observations at different azimuth angles also information on the aerosol scattering phase function can be derived. From the combined analysis of the observed intensity and the O_4 absorption also information on the single scattering albedo of the aerosol can be obtained.

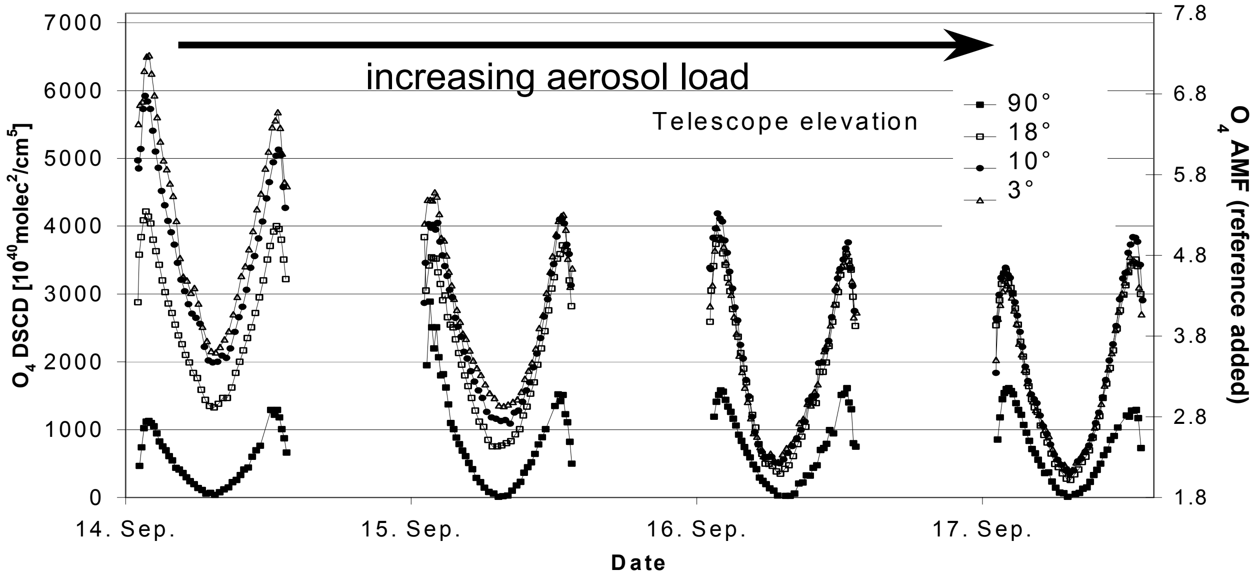

Figure 2. O_4 DSCDs measured for different elevation angles during four mostly clear days (September, 14 – 17, 2003) at Milan, Italy. Because of the changing sun position during the day the relative azimuth angles of the telescopes with respect to the sun are ~90° during sunrise, ~0° during noon and ~–90° during sunset. The main effects of aerosol scattering are the general reduction of the observed O_4 absorption and the decreasing difference between the low elevation telescopes (3°, 10°, 18°). These effects can be related to the increased aerosol load during the selected period (see text). The zenith observations during the morning of September, 15 were affected by sporadic clouds and should not be taken into account for the detailed interpretation. For the comparison of the measurements to the results of radiative transfer models the respective AMF is also shown (right axis) (Wagner et al., 2004).

MAX-DOAS observations are sensitive even to very weak aerosol optical depths. Assuming typical measurement errors we conclude that aerosol optical depths >0.001 should be clearly detectable by MAX-DOAS O_4 observations [*Wagner et al.,* 2004].

Conclusions

Compared to established methods like sun radiometers and LIDAR measurements MAX-DOAS O_4 observations have two major advantages: Since they analyze the differential O_4 absorption structures they do not require an absolute radiometric calibration or application of Langley-plot techniques. Thus they are in particular insensitive to instrument degradation. In addition, since the atmospheric O_4 profile depends strongly on altitude, MAX-DOAS O_4 observations are sensitive to the atmospheric light path distribution. Therefore, they can especially yield information on the atmospheric aerosol profile. MAX-DOAS O_4 observations at different wavelengths can characterize the wavelength dependence of the aerosol scattering.

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