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Optical Absorption
Spectroscopy**

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Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS)

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Abstract

Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) is a novel measurement technique that represents a significant advance on the well-established zenith scattered sunlight DOAS instruments which are mainly sensitive to stratospheric absorbers. MAX-DOAS utilizes scattered sunlight received from multiple viewing directions. The spatial distribution of various trace gases close to the instrument can be derived by combining several viewing directions. Ground based MAX-DOAS is highly sensitive to absorbers in the lowest few kilometres of the atmosphere and vertical profile information can be retrieved by combining the measurements with Radiative Transfer Model (RTM) calculations. The potential of the technique for a wide variety of studies of tropospheric trace species and its (few) limitations are discussed. A Monte Carlo RTM is applied to calculate Airmass Factors (AMF) for the various viewing geometries of MAX-DOAS. Airmass Factors can be used to quantify the light path length within the absorber layers. The airmass factor dependencies on the viewing direction and the influence of several parameters (trace gas profile, ground albedo, aerosol profile and type, solar zenith and azimuth angles) are investigated. In addition we give a brief description of the instrumental MAX-DOAS systems realised and deployed so far. The results of the RTM studies are compared to several examples of recent MAX-DOAS field experiments and an outlook for future possible applications is given.

1. Introduction

The analysis of the atmospheric composition by scattered sunlight absorption spectroscopy in the visible/near ultraviolet spectral ranges has a long tradition. This application is also called “passive” absorption spectroscopy in contrast to spectroscopy using artificial light sources (i.e. active DOAS, Perner et al., 1976; Platt et al., 1979; Perner and Platt, 1979; Platt and Perner, 1980; Platt et al., 1980).

Shortly after Dobson and Harrison (1926) conducted measurements of atmospheric

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ozone by passive absorption spectroscopy, the “Umkehr” technique (Götz et al., 1934), which was based on the observation of a few (typically 4) wavelengths, allowed the retrieval of ozone concentrations in several atmospheric layers yielding the first vertical profiles of ozone.

5 The COSPEC (CORrelation SPECTrometer) technique developed in the late 1960s was the first attempt to study tropospheric species by analysing scattered sunlight in a wider spectral range while making use of the detailed structure of the absorption bands with the help of an opto-mechanical correlator (Millan et al., 1969; Davies, 1970). It has now been applied for over more than three decades for measurements of total
10 emissions of SO₂ and NO₂ from various sources, e.g. industrial emissions (Hoff and Millan, 1981) and – in particular – volcanic plumes (e.g. Stoiber and Jepsen, 1973; Hoff et al., 1992).

Scattered sunlight was later used in numerous studies of stratospheric and (in some cases) tropospheric NO₂ as well as other stratospheric species by ground-based differential optical absorption spectroscopy (DOAS), as summarised in Table 1. This was
15 a significant step forward, since the quasi-continuous wavelength sampling of DOAS instruments in typically hundreds of spectral channels allows the detection of much weaker absorption features and thus higher sensitivity. Moreover the simultaneous measurement of several absorbers is possible while cross-interferences and the influence of Mie scattering are virtually eliminated.

20 Scattered sunlight DOAS measurements yield “slant” column densities of the respective absorbers. Most observations were done with zenith looking instruments because the radiative transfer modelling necessary for the determination of vertical column densities is best understood for zenith scattered sunlight. On the other hand, for studies
25 of trace species near the ground, artificial light sources (usually high pressure Xenon short arc lamps) were used in active Long-path DOAS experiments (e.g. Perner et al., 1976; Perner and Platt, 1979; Platt et al., 1979, Mount, 1992, Plane and Smith, 1995, Axelson et al., 1990, Stutz and Platt, 1997). These active DOAS measurements yield trace gas concentrations averaged along the several kilometre long light path, extend-

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ing from a searchlight type light source to the spectrometer. Active DOAS instruments have the advantage of allowing measurements to be made independent of daylight and at wavelengths below 300 nm, however, they require a much more sophisticated optical system, more maintenance, and one to two orders of magnitude more power than passive instruments (e.g. Platt, 1994). Therefore a type of instrument allowing measurements of trace gases near the ground like active DOAS, while retaining the simplicity and self-sufficiency of a passive DOAS instrument, is highly desirable.

Passive DOAS observations, essentially all using light scattered in the zenith, had already been performed for many years (see Table 1) when the “Off-Axis” geometry (i.e. observation at directions other than towards the zenith) for measurements of scattered sunlight was first introduced by Sanders et al. (1993) to observe OCIO over Antarctica during twilight. The strategy of their study was to observe OCIO in the stratosphere using scattered sunlight as long into the “polar night” as possible. As the sun rises or sets, the sky is of course substantially brighter towards the horizon in the direction of the sun compared to the zenith. Thus the light intensity and therefore the signal to noise ratio is improved significantly. Sanders et al. (1993) also pointed out that the off-axis geometry increases the sensitivity for lower absorption layers. They concluded that absorption by tropospheric species (e.g. O_4) is greatly enhanced in the off-axis viewing mode, whereas for an absorber in the stratosphere (e.g. NO_2) the absorptions for zenith and off-axis geometries are comparable. Arpaq et al. (1994) used their off-axis observations during morning and evening twilight to derive information on stratospheric BrO at mid-latitudes, including some altitude information from the change in the observed columns during twilight. At the time of the measurements of Arpaq et al. (1994) the available radiative transfer models were single scattering approximations for the off-axis viewing mode, which was sufficient for the study of stratospheric absorbers.

In spring 1995 Miller et al. (1997) conducted off-axis measurements at Kangerlussuaq, Greenland in order to study tropospheric BrO and OCIO related to boundary layer ozone depletion after polar sunrise. These authors observed at off-axis angles (i.e. angle between zenith and observation direction) of 87° and 85° , respectively, to obtain a

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larger signal due to the absorption by the tropospheric BrO fraction. No comparison was reported between off-axis and zenith sky measurements. The twilight behaviour of the slant columns was used to identify episodes of tropospheric BrO.

Off-axis DOAS, partly in combination with using the moon as direct light source, was also employed for the measurement of stratospheric and tropospheric profiles of NO₃ by ground based instruments (Weaver et al., 1996; Smith and Solomon, 1990; Smith et al., 1993; Kaiser, 1997; von Friedeburg et al., 2002).

An overview of the different DOAS measurement configurations and the respective measured quantities reported to date is given in Table 1.

Here we present a new approach to the problem of measuring tropospheric species by observing their absorption in scattered sunlight. This technique combines the advantages of all preceding attempts and introduces several new concepts: Combination of measurements at several viewing angles, multiple scattering radiative transport modelling, and the use of the O₄ absorption to quantify Mie scattering and aerosols. It allows the study of atmospheric trace gases close to the instrument (i.e. in the boundary layer with ground based instruments) with extreme sensitivity and some degree of spatial resolution. The instrument is very simple in its set-up and can be used from the ground as well as from various airborne platforms.

2. The DOAS technique

Differential Optical Absorption Spectroscopy (DOAS) is a technique that identifies and quantifies trace gas abundances with narrow band absorption structures in the near UV and visible wavelength region in the open atmosphere (e.g. Platt, 1994). The fundamental set-up of a DOAS system consists of a broadband light source, an optical set-up that transfers the light through the atmosphere, and a telescope – spectrograph – detector system to record the absorption spectra. The basic idea of DOAS is to separate the trace gas absorption cross section into two parts, one that varies “slowly” with wavelength, and a rapidly varying differential cross section σ' . The latter can be

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thought of as absorption lines or bands. Broadband extinction by Mie scattering, instrumental effects and turbulence are difficult to quantify, therefore these interferences have to be corrected to derive trace gas concentrations. If the same filtering procedure is applied to the atmospheric absorption spectrum, the narrow band absorption can be used to calculate the trace gas concentrations (Platt, 1994). The advantages of DOAS are the ability to detect extremely weak absorptions (O.D. $\sim 10^{-4}$), the unequivocal and absolute identification of the trace gases, as well as the fact that trace gas concentrations are determined solely from the absorption cross section. A calibration is therefore not necessary. DOAS uses the Beer-Lambert law with modified source intensity I'_0 and absorption cross section σ' to eliminate contributions that vary only “slowly” with wavelength:

$$I(\lambda) = I'_0(\lambda) \cdot e^{-\sigma'(\lambda) \cdot S} \quad S : \text{Slant Column Density (SCD)}. \quad (1)$$

There are several aspects which are characteristic of scattered sunlight measured by passive DOAS instruments:

2.1. The Fraunhofer reference spectrum

The solar radiation can be described, in first approximation, as the continuous emission of a black body with $T \approx 5800$ K. This continuum, however, is overlaid by a large number of strong absorption lines called the Fraunhofer lines. These lines are due to selective absorption and re-emission of radiation by atoms in the solar photosphere. Many solar Fraunhofer lines are dominant in scattered sunlight DOAS, especially in the UV and visible spectral range (300–600 nm) and are substantially stronger than absorption due to most constituents of the terrestrial atmosphere.

Fraunhofer lines have to be carefully removed in the DOAS analysis procedure in order to evaluate the absorption structures of the much weaker absorptions due to trace gases in the earth's atmosphere (optical densities of 10^{-3} and less compared to Fraunhofer lines with up to 30% absorption at typical DOAS spectral resolution). A so-called Fraunhofer reference spectrum (FRS) is always included in the fitting process

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for the MAX-DOAS evaluation of scattered sunlight spectra (for details on the DOAS fit see Stutz and Platt, 1996). This spectrum can either be a single, carefully-chosen background spectrum or a new FRS for each series of MAX-DOAS at different viewing directions. In the former case usually one fixed FRS, taken at small solar zenith angle and zenith observation geometry for minimum trace gas absorption, is used to evaluate differential slant column densities (DSCD, S_D), which are differential with respect to this FRS. So called “tropospheric difference” slant column densities Δ SCD (ΔS) are calculated from these DSCD’s by subtracting the DSCD of the zenith viewing direction containing minimum tropospheric absorptions from the respective DSCD’s of the other viewing directions:

$$\frac{I(\lambda)}{I_{\text{FRS}}(\lambda)} = e^{-\sigma'(\lambda) \cdot (S - S_{\text{FRS}})} \rightarrow S_D = S - S_{\text{FRS}} = \frac{\ln \frac{I_{\text{FRS}}(\lambda)}{I(\lambda)}}{\sigma'(\lambda)} \forall \lambda \quad (2)$$

$$\Delta S = S_{D\alpha \neq 90^\circ} - S_{D\alpha = 90^\circ}.$$

The latter approach directly yields Δ SCD’s for the used viewing directions, which can be compared to model results (see below).

$$\Delta S = S_{\alpha \neq 90^\circ} - S_{\alpha = 90^\circ}. \quad (3)$$

Here, α denotes the elevation angle (angle between the viewing direction and the horizontal direction), which is commonly used to characterise MAX-DOAS viewing directions. Since in most cases the zenith direction ($\alpha = 90^\circ$) has been used as background FRS, DSCD’s and Δ SCD’s are defined relative to the zenith direction. It should be noted that both ways of calculating Δ SCD’s yield, of course, the same results.

$$\Delta S = S_{D,\alpha \neq 90^\circ} - S_{D,\alpha = 90^\circ} = (S_{\alpha \neq 90^\circ} - S_{\text{FRS}}) - (S_{\alpha = 90^\circ} - S_{\text{FRS}}) = S_{\alpha \neq 90^\circ} - S_{\alpha = 90^\circ} = \Delta S. \quad (4)$$

2.2. The Ring effect

The Ring effect – named after Grainger and Ring (1962) – leads to a reduction of the observed optical densities of solar Fraunhofer lines depending on the atmospheric

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light path. For example, Fraunhofer lines observed at large solar zenith angles (SZA) appear weaker ("filled in") than the same lines at small SZA. Precise measurements can only be made if this effect is compensated for, otherwise complete removal of Fraunhofer lines by division of spectra taken at small and large SZA, respectively, is impossible. Rotational Raman scattering is thought to be the most probable cause for the Ring effect (Bussemer, 1993; Fish and Jones, 1995; Burrows et al., 1996; Sioris et al., 1999). Optical density changes due to the Ring effect are of the order of a few percent, which significantly affects DOAS measurements of scattered radiation. Thus a very accurate correction is required, since the atmospheric absorptions which are evaluated are sometimes more than an order of magnitude smaller than the filling in of the Fraunhofer lines. Therefore a so called Ring reference spectrum is included in the DOAS fitting process when scattered sunlight spectra are evaluated. Fit coefficients derived for the Ring effect in MAX-DOAS measurements can for example be used to detect light path enhancements due to multiple scattering and possibly investigate other aspects of the radiative transfer.

For a more comprehensive discussion of effects that have to be addressed when using scattered sunlight for DOAS measurements see also (Solomon, 1987b; Platt et al., 1997).

2.3. The O_4 spectrum

The oxygen dimer O_4 is another important parameter for MAX-DOAS. O_4 is also referred to as $(O_2)_2$ to point out that it is a collisional complex rather than a bound molecule. Absorption bands of O_4 occur at several wavelengths in the UV/Vis spectral range (Perner and Platt 1980). Therefore an O_4 reference spectrum (Greenblatt et al., 1990) is often included in the DOAS analysis. O_4 has been used in many cases to characterize the effects of multiple scattering in clouds and other aspects of the radiative transfer on DOAS measurements of scattered sunlight (Erle et al., 1995; Wagner et al., 1998; Pfeilsticker et al., 1998; Wagner et al., 2002). For MAX-DOAS measurements O_4 can be used to determine the importance of multiple scattering and to derive

aerosol profile information (see Sect. 4, sensitivity studies, below).

3. MAX-DOAS (geometric approach)

The calculation of average trace gas concentrations from slant column density measurements using direct sunlight or active DOAS arrangements is straightforward. In the case of the analysis and interpretation of DOAS measurements using scattered sunlight, where the radiation can travel along multiple paths, it is crucial to correctly describe the radiative transfer in the atmosphere (Marquard et al., 2000). The apparent absorption of trace gases with distinct vertical profiles (e.g. O₃, NO₂, BrO) measured by a ground based spectrometer depends strongly on the distribution of the paths taken by the registered photons on their way through the atmosphere. DOAS measurements using scattered sunlight yield apparent slant column densities (SCD) S , which are defined as the trace gas concentration integrated along the effective light path (in reality it is an average of an infinite number of different light paths).

$$S = \int_0^L c(s) ds \quad \text{for direct light: } \bar{c} = \frac{S}{L}, \bar{c} : \text{average concentration.} \quad (5)$$

Note that for a single SCD measurement the individual photons registered in the detector have travelled different paths through the atmosphere before being scattered into the DOAS telescope. Therefore Eq. (5) can only account for the most probable path defined as an average over a large number of registered photons. Since the SCD depends on the observation geometry and the current meteorological conditions, it is usually converted to the vertical column density (VCD) V , which is defined as the trace gas concentration $c(z)$ integrated along the vertical path through the atmosphere:

$$V = \int_0^{\infty} c(z) dz. \quad (6)$$

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The concept of air mass factors has been used for interpreting zenith-scattered light DOAS observations for many years (e.g. Solomon et al., 1987b; Perliski and Solomon, 1993). The air mass factor (AMF) A is defined as the ratio of SCD and VCD:

$$A(\lambda, \vartheta, \alpha, \phi) = \frac{S(\lambda, \vartheta, \alpha, \phi)}{V}, \quad (7)$$

where ϑ denotes the solar zenith angle (SZA), α the telescope elevation angle and ϕ the relative azimuth angle, which is defined as the azimuth angle between the telescope direction and the sun. Taking into account the distribution of different light paths leads to an improved airmass factor concept (Marquard et al., 2000).

The observation geometry and the respective angles are shown in Fig. 1. For simplicity the relative azimuth angle ϕ is assumed to be 180° here. The AMF depends on the radiative transfer in the atmosphere and is therefore also influenced by the profiles of several parameters including trace gas concentration, pressure, temperature, profiles of strong absorbers (e.g. ozone) and aerosols (including clouds) as well as the surface albedo.

Taking a as the fraction of the total vertical trace gas column V residing below the scattering altitude we obtain the SCD (in the single scattering approximation sketched in Fig. 1):

$$S \approx \left[a \cdot \underbrace{\frac{1}{\sin \alpha}}_{A_{\text{lower layer (BL)}}} + (1 - a) \cdot \underbrace{\frac{1}{\cos \vartheta}}_{A_{\text{upper layer (strat.)}}} \right] \cdot V \quad (a \leq 1). \quad (8)$$

Therefore, an absorber near the ground (e.g. in the boundary layer) enhances the airmass factor according to approximately a $1/\sin \alpha$ relation, expressing the strong dependence of the AMF on the elevation angle α . In contrast, the airmass factor strongly depends on the solar zenith angle ϑ with approximately a $1/\cos \vartheta$ relation for an absorber in the higher atmosphere (e.g. in the stratosphere).

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4. Radiative transfer calculation of MAX-DOAS airmass factors

A set of trace gas column measurements performed at a series of different elevation angles can be used to infer the vertical trace gas profile and thus the absolute concentration of the trace gas as a function of altitude. In addition, the aerosol optical density can be quantified from the variation with the observation geometry of the SCD of species with known concentration (such as O_2 and O_4). In order to characterise the role of various parameters sensitivity studies are presented here.

As mentioned above, the geometric, single scattering approach using the equation $A \sim 1/\cos\vartheta$ (for scattering below the trace gas layer), where ϑ is the solar zenith angle, or $A \sim 1/\sin\alpha$ (for scattering above the trace gas layer), where α is the telescope elevation angle, can only be regarded as an approximation. In particular factors influencing the result like Mie scattering, ground albedo, or arbitrary distributions of trace gases can only be quantified by radiative transfer calculations. In the following we present a study to illustrate some of the properties of the MAX-DOAS technique.

Precise air mass factors for this study were calculated using the Monte Carlo radiative transfer model (RTM) "Tracy" (von Friedeburg, 2003) which includes multiple Rayleigh and Mie scattering, the effect of surface albedo, refraction, and full spherical geometry. It relies on input data including pressure and temperature profiles, ground or cloud albedo, aerosol profiles and aerosol scattering phase functions as well as profiles of strong molecular absorbers like ozone. Air mass factors and other parameters like the number of scattering events or the altitude of the last scattering event are then calculated for various assumed profiles of the respective absorbers.

The radiative transfer may vary significantly for different angles α , ϑ or ϕ , therefore radiative transfer calculations were performed to quantify the influence of the above parameters. In particular the behaviour of AMF's as a function of elevation angle, solar zenith angle, solar azimuth angle, ground albedo, and aerosol load was studied for several idealised trace gas profiles (see Table 2 and left panel of Fig. 2). We assumed constant trace gas concentrations c in the 0–1 km and 0–2 km layers of the

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atmosphere for the first four profiles (designated P1 through P4). We also investigated four further profiles for comparison, one of oxygen dimers (O_4) with $c \propto c(O_2)^2$ (P5) and a stratospheric profile (P6, centred at 25 km with a FWHM of 10 km) as well as two free tropospheric profiles (c constant at 5 ± 1 km and 8 ± 1 km, respectively).

5 The dependence of the AMF for an absorber in the stratosphere and the comparison with an absorber confined to the boundary layer has already been discussed by Hönninger and Platt (2002) for the example of bromine oxide (BrO) in the Arctic boundary layer. The dependence on the solar azimuth angle ϕ was shown to be small both, for the stratospheric as well as for the boundary layer case. A significant dependence
10 appears only for high solar zenith angles ($\vartheta > 80^\circ$).

The air mass factors in the sensitivity studies described below were calculated for the following set of parameters: Wavelength $\lambda = 352$ nm, Telescope elevation angles $\alpha = 2^\circ, 5^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 60^\circ, 90^\circ$, relative azimuth $\phi = 90^\circ$, except for the study of the azimuth dependence, solar zenith angle $\vartheta = 30^\circ$, except for the study of the SZA
15 dependence. The vertical grid size h for the horizontal layers was $h = 100$ m between 0 and 3 km, $h = 500$ m between 3 and 5 km and $h = 1$ km above 5 km up to the top of the model atmosphere at 70 km. A standard atmospheric scenario (US standard atmosphere) for temperature, pressure and ozone was used and interpolated to match the vertical grid of the model.

20 In order to characterise the role of aerosols, the following scenarios were modelled: Above 10 km a standard aerosol load from a chemical model scenario (Hendrick, pers. comm. 2003) with $\varepsilon_M \sim 10^{-3} \text{ km}^{-1}$ at 10 km was selected. Below 10 km, where the aerosol load becomes important for tropospheric absorbers due to its influence on the radiative transfer, 3 cases were chosen:

- 25 1. “Pure Rayleigh” case, no tropospheric aerosols.
2. “Low aerosol” load, with an extinction coefficient at 0 km altitude of $\varepsilon_M = 0.1 \text{ km}^{-1}$, and linear interpolation of $(\log \varepsilon_M)$ between the ground and the stratosphere (see Fig. 2, centre panel).

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3. “High aerosol” load with extinction coefficient at 0 km of $\varepsilon_M = 1 \text{ km}^{-1}$, and linear interpolation of $(\log \varepsilon_M)$ between the ground and the stratosphere.

4. Two scattering phase functions were distinguished for the cases 2 and 3: (a) typical continental aerosol and (b) typical marine aerosol (see Fig. 2 right panel).

5 The single scattering albedo of the aerosols was set to 1 for simplicity. All model runs were performed for two ground albedo values of 5% and 80%, respectively.

4.1. The dependence of the AMF on the observation geometry

In order to demonstrate the sensitivity of MAX-DOAS towards various vertical trace gas profiles we first focus on a simple atmospheric scenario and discuss the dependence of the AMF on the viewing direction (parameterised as elevation angle α) for several vertical trace gas profiles. In Fig. 3 the modelled air mass factors for the profile shapes P1–P6 are plotted as a function of the viewing direction for the pure Rayleigh case and low (left) and high (right) ground albedo.

The AMF is strongly dependent on the viewing direction (elevation angle α) for the profiles, where the absorber is close to the ground. The AMF for the profiles extending to the ground (P1, P2, P3, P5) increases continuously when the elevation angle decreases. This effect can still be understood in the geometric approximation discussed in Sect. 3. The viewing direction determines the absorption path length, which decreases continuously for P1, P2, P3 and P5 when α increases. The increase is strongest for the lowest profile P1 and less strong for the profiles extending to higher altitudes. The maximum AMF for the elevated layer profile P4 appears at $\alpha = 5^\circ$ and AMF decreases for smaller and larger α . Simply speaking the instrument looks “below” the layer at $\alpha < 5^\circ$. In contrast to that the AMF is almost independent of the viewing direction for the stratospheric profile P6. The characteristic altitude that determines how the AMF depends on the viewing direction is the mean last scattering altitude (LSA, see below).

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4.2. Aerosol and albedo dependence

Aerosol load and scattering properties as well as the surface albedo play a significant role in the radiative transfer, especially in the lower troposphere. In Fig. 4 the AMF dependence on the observation elevation angle is shown again for the different profile shapes (P1–P6), this time not for the pure Rayleigh troposphere, but for two different aerosol scenarios (high and low tropospheric load) as well as for high and low ground albedo. The calculations assuming low aerosol load are not different in general shape from the Rayleigh case, however the absolute values of the AMF are considerably smaller at elevation angles of 10° and lower. In contrast, at high aerosol load the increase of the AMF at lower elevation angles becomes much weaker, and the variation of the AMF with elevation angle becomes much less pronounced.

The air mass factors at low elevation angles are generally smaller for increased aerosol load compared to the Rayleigh case, while they increase at high elevation angles. However the increase of the AMF with smaller elevation angles remains very similar for the profiles extending to the ground (P1–P3) in the low aerosol load scenario. It can already be seen in the low aerosol load scenario, that the AMF is strongly decreasing (compared to the Rayleigh case) for the profiles extending to higher levels, e.g. P4 and P5. This appears to be due to the shorter mean free path of photons due to the aerosol extinction. Therefore, the absorption path in the lowest atmospheric layers is shorter in these cases and a more vertical path in the higher layers is probable as is sketched in Fig. 5. This also explains why deviations from the AMF behaviour based on simple geometric assumptions (as sketched in Fig. 1) become more and more obvious for higher aerosol loads.

This effect is even stronger for the high aerosol load scenario due to shorter mean free paths and correspondingly lower scattering altitudes near the ground. The maximum of the AMF curve can shift towards higher elevation angles by as much as 40° (e.g. the lifted layer profile P4 in Fig. 4, high aerosol, 80% ground albedo case). However, shorter mean free paths also lead to a higher average number of scattering events

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(see Fig. 6), thus reducing the differences between the various viewing directions in a way, that differences might not be significant anymore in measurements.

Two different aerosol phase functions, continental and maritime (see Fig. 2) were investigated, results are shown in Fig. 7. The phase function for the marine aerosol (Fig. 2) indicates slightly favoured forward scattering compared to the continental case. However, no significant differences between the two aerosol types can be seen here, since the phase functions are quite similar.

The effect of higher albedo generally enhances the AMF for the profiles extending to the ground (P1–P3), because longer absorption paths in the lowest layers are favoured by scattering at the ground. Since the effect of the higher albedo increases all AMF's by almost the same amount, it has no significant effect on the elevation angle dependencies, and rather constitutes an offset.

As mentioned above, another parameter that determines the sensitivity of MAX-DOAS towards absorbers with different profile shapes is the number of scattering events that photons undergo on average prior to being detected by the MAX-DOAS instrument. Figure 6 shows the contribution of the different scattering processes as a function of elevation angle for the typical low aerosol scenario for both low (5%) and high (80%) ground albedo.

It can clearly be seen that a higher ground albedo strongly increases the overall number of scattering events compared to the low albedo case. The probability of Mie and ground scattering processes increases, especially for the low elevation angles. Since both scattering processes take place largely at or close to the ground, this finding is not surprising. However, the specific behaviour and the influence on the respective air mass factors can only be modelled properly using an RTM. The albedo increase from 5% to 80% has also a significant effect on the number of Rayleigh scattering events, since more light reaching the ground is reflected and can undergo more scattering processes.

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4.3. The Last Scattering Altitude (LSA)

A key parameter that governs the sensitivity of MAX-DOAS measurements towards different vertical profiles of absorbers is the altitude of the last scattering event (last scattering altitude, LSA, see also Fig. 5) before the photon finally reaches the MAX-DOAS instrument. The LSA dependence on the viewing direction is shown in Fig. 8. As expected, the LSA becomes smaller for the lower elevation angles. This can lead to the observed behaviour of the AMF with a maximum between 10° and 20° , and smaller AMF for the very low elevation angles (e.g. for P3). The LSA effectively determines the length of the last section of the path taken by the photons through the lowest atmospheric layer at the observation elevation angle α .

This analysis explains the following effects:

- For profiles extending to the ground and with vertical extensions comparable to the LSA, such as P1–P3, the AMF is generally smaller than estimated from the geometrical ($1/\sin\alpha$)-approximation.
- For profiles elevated from the ground and/or with a significant fraction of the profile above the respective LSA, such as P4 or P5, the AMF has a maximum at an elevation angle, where the mean free path still allows transecting the absorbing layer on a long slant path. AMF's decrease again for the very small elevation angles due to light paths being shorter in the layers with high absorber concentrations.
- For stratospheric profiles, such as P6, the last scattering processes near the ground cannot influence the AMF since the absorber is only present at high altitudes. Therefore, the AMF for stratospheric absorbers is largely independent of the viewing direction.

4.4. “Box” air mass factors

Box AMF's are a measure of the light path enhancement compared to a vertical path in a given height interval. In the case of weak absorption and a horizontally homogeneous

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atmosphere the Box AMF can be expressed as:

$$A_{\text{Box}} = \frac{\int_{\text{Box}} ds}{\int_{\text{Box}} dz}, \text{ with the actual light path } s \text{ and the vertical path } z. \quad (9)$$

Box Air Mass Factors modelled for different viewing directions (elevation angles) therefore serve as a measure of the sensitivity of a particular viewing direction towards an absorber being present in a specific vertical “box” (or layer). Therefore, Box AMF’s provide a criterion for determining the sensitivity of MAX-DOAS for different shapes of vertical profiles.

Figure 9 shows modelled Box AMF’s for assumed trace gas layers of 100 m thickness located below 2 km altitude. Variations of the Box AMF’s are clearly strongest towards the surface for the very low elevation angles. Consequently MAX-DOAS should be very sensitive to different profile shapes in this altitude range. The general trend for elevation angles smaller than 5° of increasing Box AMF with decreasing α is only observed for layers below 400 m altitude. Above this altitude, the trend reverses and Box AMF’s decrease.

This confirms that a good knowledge of the aerosol load of the probed airmass is important in order to choose the correct AMF and thus to derive quantitative trace gas vertical columns V . In fact, some information about the vertical trace gas distribution can be gained from an analysis of the variation of the observed slant column with the elevation angle, as explained above; the vertical thickness D of a trace gas layer can be derived. This information can be used to infer the average trace gas concentration in the layer.

4.5. O_4 profile and measurements as good indicator for the aerosol load

The observation of the slant columns of oxygen dimers (and other species with known atmospheric distribution, e.g. O_2) offers an excellent opportunity to determine the

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aerosol load of the atmosphere. The atmospheric O_4 concentration profile is well known; it is essentially given by $c_{O_4}(z) \propto c_{air}^2(z)$ with a known dependency of the air density c_{air} on temperature and barometric pressure. The measured set of O_4 slant column densities (for a series of elevation angles e.g. from 2° to 90°) is then compared to a series of calculated O_4 SCD's (for the temperature and pressure profile as recorded during the measurement), where the aerosol load is varied until best agreement is reached. As outlined in Fig. 10 this aerosol load is then used to calculate a further series of "Box" AMF's which, together with the slant column densities of the trace gases under investigation, are used as input for a linear equation system. Alternatively, the AMF for several test profiles are calculated and the "best fit" vertical trace gas profile (of e.g. the type P1...P4) is chosen.

4.6. Azimuth dependence of the AMF

A dependence of the air mass factors on the relative azimuth angle between the sun and the viewing direction can arise from the shape of scattering phase functions for the respective scattering processes. Therefore, light paths taken by photons at different relative azimuth angles may be different resulting in different elevation angle dependence of the AMF and possible results on MAX-DOAS measurements. This effect is expected to be higher for large SZA since only then can one expect significant influence on the radiative transfer at low altitudes which is most important for MAX-DOAS measurements (Hönninger and Platt, 2002).

Figure 11 shows the small effect of the relative azimuth angle for a SZA of 30° and the profile P4. No significant influence on the general elevation angle dependence of the AMF is seen. However, the small azimuth dependence is found to increase with ground albedo and tropospheric aerosol load.

4.7. SZA dependence of the AMF

The solar zenith angle (SZA) is the most important parameter when characterizing air mass factors for stratospheric absorbers. In Fig. 12 the strong dependence of the AMF on the SZA can be seen for the stratospheric profile P6, whereas the viewing direction only plays an insignificant role. In contrast to that for the tropospheric profiles (e.g. P2 and the O₄ profile P5) the SZA dependence of the AMF is minor and only significant at large SZA. Here, the viewing direction (elevation angle) is the parameter that governs the AMF as discussed above.

The SZA dependence of the AMF for the different profiles can be best understood from the altitudes of the first and last scattering event (FSA, LSA, respectively). These parameters can serve as a proxy for the general pattern of the light path taken by the registered photons. Figure 13 shows both first and last scattering altitude as a function of SZA. While the FSA strongly increases towards high SZA, the LSA is largely independent of SZA. As long as the FSA is below or still within an absorber profile (which is the case for the O₄ profile P5 up to a higher SZA than is the case for P2), the first light path segment (above FSA) increases strongly with SZA (approximately according to $1/\cos(\text{SZA})$). As soon as the FSA is above the absorber, the first slant path segment does not increase the AMF and therefore the AMF dependence on the SZA disappears.

4.8. Profiles of free tropospheric absorbers

As discussed above, ground-based MAX-DOAS can clearly distinguish between stratospheric profiles (e.g. P6) and various tropospheric profiles (P1–P5). However, the limitation of the sensitivity towards profiles at higher altitudes remains to be investigated. Figure 14 shows the SZA dependence of the AMF for two layers with constant trace gas concentration in the free troposphere (P7 and P8, see Table 2 for definition).

The SZA dependence of the profiles P7 and P8 is not as strong as for the stratospheric profile P6, but clearly more visible than for the boundary layer profiles or the

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O₄ profile P1–P5. On the other hand, the differences between the various elevation angles (shown as different colours in Fig. 14) are much smaller than for the lower tropospheric profiles. Differences for various elevation angles are still significant in MAX-DOAS measurements for a free tropospheric profile like P7, however, for profile P8 the small differences between the viewing directions are even more difficult to measure and to distinguish from measurement errors, modelling noise and from the variability of the trace gas concentration.

5. Practical realization of MAX-DOAS systems

As described above MAX-DOAS systems have been employed for the measurement of atmospheric trace gases by several authors (see Table 1). A number of technical requirements have to be fulfilled by MAX-DOAS instruments. Besides sufficient spectral resolution (around 0.5 nm in the near UV) in particular a sufficiently small field of view of the telescope (typically < 1°) is required, and of course the instrument must allow measurements in different viewing directions. A number of technical approaches have been developed to meet these requirements:

1. Sequential observation at different elevation angles. This approach has the advantage of a relatively simple set-up requiring only one spectrometer and some mechanism for pointing the telescope (or the entire spectrometer-telescope assembly) in the desired directions. It was therefore used in several studies (Hönninger and Platt, 2002; Leser et al., 2003; Bobrowski et al., 2003; Hönninger et al., 2003a; Hönninger et al., 2003b) Disadvantages are that the measurements are not simultaneous, typically a complete cycle encompassing several (usually 4 to 10) observation directions may take 5 to 30 min (e.g. Hönninger and Platt, 2002). While the effect of changing SZA and thus stratospheric trace gas columns can be overcome by interpolation (Leser et al., 2003), this can be a problem during periods of rapidly varying radiative transport conditions in the atmosphere e.g.

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varying cloud cover, aerosol load or during sunrise/sunset. Also the instrument contains mechanically moving parts, which may be a disadvantage in long term operation at remote sites. Very robust, lightweight and small-sized Mini-MAX-DOAS instruments have recently been applied for automated operation at remote sites (Galle et al., 2003; Bobrowski et al., 2003; Hönninger et al., 2003b) (see Fig. 15).

2. Simultaneous observation at different elevation angles allows truly simultaneously determination of SCD's at the different elevation angles and thus avoids the problems of set-up 1. Disadvantages here are the larger instrumental requirement, since a spectrometer and a telescope for each of the observation directions is needed. However recent development of very compact, low cost spectrometer/detector combinations (e.g. Galle et al., 2003; Bobrowski et al., 2003; see Fig. 15) can be of help here. Alternatively some simplification may come from using spectrometers with two dimensional detectors, (see Fig. 16 and description below) (Heismann, 1996; Wagner et al., 2002; Löwe et al., 2002; Bossmeyer, 2002; Oetjen, 2002; von Friedeburg, 2003; Heckel, 2003).

A severe problem arises, however, if the different instruments are to be ratioed against each other in order to eliminate the solar Fraunhofer structure (see Sect. 2). This proves extremely difficult if different instruments with – in practice – different instrument functions are involved (e.g. Bossmeyer, 2002; von Friedeburg, 2003). Although the situation can be improved by “cross convolution” (i.e. convolving each measured spectrum with the instrumental line shape of the respective spectrum it is to be compared with), or by generating difference spectra from FRS, it appears that, given present instrumentation, the highest sensitivity is reached with sequential observation.

3. A solution lies in the combination of techniques 1) and 2), i.e. the use of multiple spectrometers (one per observation direction) and moving telescopes (or spectrometer telescope assemblies). While this approach appears to combine

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the disadvantages of the two above set-ups it also combines their advantages: If each individual instrument sequentially observes at all elevation angles and the observations are phased in such a way that at any given time one instrument observes each observation direction, then indeed not only will simultaneous observation be achieved at each angle, but also each instrument regularly sees the zenith to record a Fraunhofer spectrum for reference. Instruments based on this approach are presently used on cruises of the German research vessel Polarstern (see Leser et al., 2003; Bossmeyer, 2002).

5.1. Spectrometer with two dimensional detector for simultaneous MAX-DOAS observation

In the simultaneous MAX-DOAS version light from several telescopes collecting scattered sunlight at the desired elevation angles is transmitted to the entrance slit of the spectrometer by quartz fibres (see Fig. 16). In order to separate the spectra of the light observed at the different viewing angles two-dimensional CCD arrays are needed as detectors (Heismann, 1996). The light is dispersed in λ (x)-direction (Fig. 16) and the different spectra for the light from different fibres are separated in y-direction (with each spectrum covering several rows of the CCD array). Thus it is possible to measure the different spectra simultaneously. Unfortunately the instrumental transmission function for the individual viewing directions may not be sufficiently similar to allow ratioing to the zenith, thus two dimensional spectrometers are best combined with technique 3 (see above).

6. MAX-DOAS observations

6.1. Comparison of measured and modelled SCD's

As discussed above, RTM calculations yield airmass factors for a priori trace gas profiles with the elevation angle as important parameter. This permits the derivation of

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vertical profile information from actual MAX-DOAS measurements. DSCD's, which result from the MAX-DOAS analysis for different elevation angles, are usually differential SCD's with respect to a Fraunhofer reference spectrum taken at the zenith viewing direction. In order to eliminate possible bias Δ SCD's are either directly derived by choosing a spectrum taken simultaneously or within short time as Fraunhofer reference for the DOAS analysis. This procedure yields Δ SCD's, which can also be calculated from DSCD's (when only a single FRS was taken for the entire DOAS analysis) as shown in Sect. 2 above.

Here $\alpha = 90^\circ$ was assumed for recording the FRS, however, in special cases a different viewing direction could be chosen for the FRS.

Figure 17 shows an example for measurements taken during the Polar Sunrise Experiment Alert 2000 by Hönninger and Platt (2002). Measured data points (Δ SCD's) are plotted as a function of the elevation angle. Additionally the model predicted Δ SCD's calculated from the boundary layer column $BL\text{-}VCD = 2 \times 10^{13} \text{ cm}^{-2}$ and the differential AMF $\Delta AMF(\alpha) = A(\alpha) - A(\alpha = 90^\circ)$ are shown for the model layers P1-P6. The measured values agree well with the model results for P1, representing a constant trace gas profile over the lowest 1 km of the atmosphere. The other profiles are not compatible with the measured data in this case. It can also be seen that the most significant differences between the modelled Δ SCD's for the various profiles appear at the low elevation angles. For deriving vertical profile information using MAX-DOAS, measurements at low elevation angles are therefore crucial.

First MAX-DOAS observations of BrO in the Arctic boundary layer (Alert, 82.5° N) during spring of 2000 were already reported by Hönninger and Platt (2002). Here we show a more comprehensive data set including stratospheric absorbers and O_4 . Figure 18 shows the MAX-DOAS results (time series of 3 weeks), where the change in solar zenith angle is very small compared to mid-latitudes (diurnal Δ SZA = 15°). O_3 and NO_2 show the typical pattern of stratospheric absorbers, which only depend on the SZA. Changes in tropospheric O_3 are masked by the large stratospheric O_3 column. The vertical ozone column changes only by about 1% in the high Arctic during

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polar sunrise, where the whole boundary layer ozone column is destroyed during frequent episodes of boundary layer ozone depletion. With the AMF for the stratospheric ozone column in the range of 5 at SZA=80°, even the high sensitivity of MAX-DOAS at the surface is not sufficient to measure this effect of typically 2–3%. In the NO₂ time series several pollution events (21, 22, 26 April, 7 May) can be identified as low elevation angle data points deviating from the general pattern which tracks the SZA change. These episodes are always correlated to exhaust plumes being blown into the MAX-DOAS viewing direction which originated either from a nearby generator or from the Alert base at several kilometres distance. On 27 April 2000 BrO ΔSCD's rise from background levels of < 10¹³ molec/cm² up to 10¹⁵ molec/cm² and simultaneously large differences can be seen between the used MAX-DOAS elevation angles. Hönninger and Platt (2002) inferred a BrO layer of roughly 1 km thickness at the surface from the MAX-DOAS results at 4 different elevation angles using only single scattering RTM calculations. Episodes of drifting snow (e.g. on 27/28 April) can also be modelled with the new RTM by von Friedeburg (2003). Also a more comprehensive investigation of the evolution of the BrO layer over time is possible (von Friedeburg, 2003). O₄ shows very little diurnal variation but the typical change in absorption with changing elevation angle for a lower tropospheric absorber. Effects of multiple scattering can be seen for the drifting snow period between 26 and 29 April (Hönninger, 2002). The measured column densities are elevated for all viewing directions as a result of enhanced light path lengths in the lowest atmospheric layer. Additional information on the aerosol profile can be derived by applying the approach mentioned above (see Sect. 4, sensitivity studies O₄).

Another example illustrating the effects of clouds on the radiative transfer and the detection by MAX-DOAS is shown in Fig. 19 for measurements at mid-latitudes. Ship borne MAX-DOAS measurements of BrO, NO₂ and O₄ were performed during a cruise of the German research vessel FS Polarstern from Bremerhaven, Germany to Cape Town, South Africa (Leser, 2001; Leser et al., 2003). A major difference in comparison with the above example at high latitudes is the range of the solar zenith angle over the

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course of the day. In Fig. 19 the strong influence of the change in SZA on the MAX-DOAS measurements can be seen as U-shaped diurnal pattern. Leser et al. (2003) presented a method to account for the change in SZA during sequential MAX-DOAS measurements by fitting a polynomial function to the diurnal DSCD pattern and calculating ΔSCD 's using interpolated values for SCD_{FRS} . This method was applied successfully for measurements at mid-latitudes by Leser et al. (2003). On the partly cloudy day shown in Fig. 19 (left) enhanced DSCD's due to light path enhancement in clouds are seen for the lowest elevation angles (5° , 10° and 20°).

Another application of ground-based MAX-DOAS is the study of tropospheric pollutants like NO_2 and SO_2 . During the ship borne MAX-DOAS measurements by (Leser, 2001; Leser et al., 2003) it was also possible to study pollution episodes. Figure 20 demonstrates the sensitivity of MAX-DOAS to NO_2 in the boundary layer in the North Sea. During enhanced NO_2 levels in the boundary layer the typical pattern of strongly increasing DSCD with decreasing elevation angle is found on 1 October 2000 (Fig. 20, left panel) when the ship cruised in the North Sea. On 12 October 2000 a typical U-shaped diurnal pattern of DSCD's with no significant differences between the measurements at various elevation angles shown in Fig. 20 (right panel) represents stratospheric background NO_2 and an unpolluted troposphere.

7. Comparison of passive MAX-DOAS with active long path-DOAS

During a field campaign at the south-east shore of the Hudson Bay, Canada, simultaneous measurements of BrO using an active Long path-DOAS system (e.g. Platt 1994) and a passive MAX-DOAS instrument were performed. The active long path DOAS yielded average BrO concentrations over a 7.6 km light path at ~ 30 m altitude above the sea ice surface. The passive MAX-DOAS was also set-up on a hill ~ 30 m above the sea ice. It used elevation angles of 5° , 10° , 20° and 90° above the horizon and the viewing azimuth was true north, less than 10° relative to the long path DOAS light path, so the same airmass was sampled in both cases (Hönninger et al., 2003a).

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An example of the intercomparison results is shown in Fig. 21. From noon to sunset the BrO as measured by the long path DOAS system showed high BrO values of 18 ppt BrO at noon decreasing to < 5 ppt in the evening. The MAX-DOAS BrO Δ SCD's are shown in the lower part of Fig. 21 for the elevation angles of 5°, 10° and 20°.

5 Around 16:00 h the elevation angle dependence compares best with a ~1 km BrO layer at the surface. Therefore, the 5° Δ SCD's were converted to mixing ratios using this assumption. Indeed, the absolute values of the MAX-DOAS mixing ratios (black data points and line in Fig. 20, bottom panel) match the Long path-DOAS data (top panel of Fig. 20) very well. Small differences can be explained by variability of the
10 boundary layer thickness. It can also be seen that the time resolution of the MAX-DOAS instrument is comparable to and often better than that of the active DOAS (Hönninger et al., 2003a).

8. Other MAX-DOAS applications

Besides the basic MAX-DOAS geometries described above many additional variants
15 are possible, which have specific advantages for the measurement in particular situations. In the following we give a few examples:

8.1. MAX-DOAS from elevated standpoints

One possible application of the MAX-DOAS approach is to make observations from
20 elevated standpoints, e.g. towers, mountains, balloons, or aircraft, which allows studies of:

- Emission patterns of source areas near the instrument,
- vertical profiles of trace gases on local and regional scales,
- free tropospheric background conditions and pollution episodes in Alpine environments,

- smog episodes in valleys which are isolated from the free troposphere by temperature inversions.

A possible MAX-DOAS experiment to study halogen release processes associated with the surface of a salt flat is shown in Fig. 22. High albedo surfaces of salt (also snow, sea surface glint, etc.) increase the light intensity and thereby the sensitivity of the measurements. In the sketched case background absorption can be easily determined and corrected for in order to yield slant column densities in the layer between the MAX-DOAS altitude and the salt lake surface. The spherically averaged absorption signal for the atmosphere above the MAX-DOAS site can be determined by analysing scattered and direct sunlight collected by a 2π integrating optics (e.g. known from measurements of down welling actinic fluxes by filter- and spectroradiometers, Junkermann et al., 1989). The light collected by this device is representative for the light which is then diffusely reflected by the high albedo salt surface. Measuring only the direct sunlight would not be appropriate since in the blue and UV spectral ranges a significant fraction of the down welling solar radiation is scattered sunlight, even under clear sky conditions ($\sim 80\%$ at 300 nm, $\sim 40\%$ at 420 nm, Lefer et al., 2001). Thus one MAX-DOAS telescope should measure this signal, which can then be used to correct slant column densities derived from the various geometries observing different locations of the bright surface below the instrument. Spatially resolved trace gas concentrations can be derived, depending on the scale of inhomogeneities, by applying analytical or numerical inversion techniques. This information can improve the knowledge of reactive halogen release processes, known to occur on sea ice or salt surfaces (Hebestreit et al., 1999; Platt, 2000; Stutz et al., 2002; Hönninger et al., 2003b). Possible local inhomogeneities including vertical profiles can be studied by MAX-DOAS observations. In addition artificial high reflectivity surfaces may be used. For instance, a quite well-defined absorption path would result from the measurement, from several kilometres away, of reflected light, from a patch of high reflectivity (e.g. a SPECTRALON sheet) of size a square meter or so. A quasi-long path absorption measurement is possible with this set-up.

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The background signal, containing absorption from the higher tropospheric layers and the stratosphere, can be eliminated by correction with zenith sky absorption measurements. Vertical profiles can be measured by pointing the light receiving telescopes at bright surfaces (e.g. snow or ice) at different altitudes. Pollution in valleys can be monitored from mountains by observing light scattered back from the usually cloudy top of the inversion layers which frequently isolate the air in the valleys. Multiple scattering in the valley leads to large absorption paths and absorption signals in the observed backscattered light. Besides being useful for simple, qualitative assessment of trace gas distributions, this method can also be used to calculate concentrations by ratioing to the simultaneously measured O₄ absorption, which is proportional to the effective light path.

A quite flexible deployment strategy is to mount the spectrometer on an aircraft (e.g. Wagner et al., 2002; Wang et al., 2003; Heue et al., 2003) known as Airborne MAX-DOAS (AMAX-DOAS). Other possible platforms include remotely piloted aircraft, airships (“Zeppelin” type dirigibles) and balloons.

Note that the assumption of a straight well-defined light path between the observed high albedo surface and the MAX-DOAS instrument is valid for many cases due to the relatively long mean free path of photons at higher altitudes (>21 km for 360 nm at 3000 m altitude).

8.2. Industrial, urban, biomass burning and volcanic plume monitoring

Routine monitoring of trace gases involved in urban air pollution is also a possible application for the MAX-DOAS technique. While the method does not yield localized concentration data like point measurements from in situ monitors, it is very sensitive to major pollutants (e.g. NO₂, SO₂, HCHO, etc.) and average concentrations in the boundary layer can be derived. For instance MAX-DOAS measurements of NO₂ near a highway were described by von Friedeburg (2003). In fact, average concentrations often prove to be much more useful than highly variable, in situ measurements which have to be averaged over time. This is particularly true when characterizing the levels

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of pollution in cities and urban areas. Sensitivity studies using air-mass factor modelling and actual measurements performed by Hönninger and Platt (2002) show that MAX-DOAS measurements alone provide sufficient information to derive the approximate height of the mixing layer and calculate average mixing ratios. Certainly in highly polluted environments where variable visibility is a major problem for other optical monitoring techniques (e.g. long path DOAS) MAX-DOAS measurements are better suited because the method also works at low visibility. With the air-mass factor concepts and in particular using information from simultaneous O_4 absorption measurements as described in this paper, quantitative measurements during cloudy and low visibility periods are also possible. Apart from scattered sunlight, the high albedos of certain points in urban areas or even guided mirrors to observe reflected sunlight provide another potential source of MAX-DOAS observation geometries.

An instrumental set-up of MAX-DOAS for the study of volcanic (Galle et al., 2003; Bobrowski et al., 2003) or other emission plumes is sketched in Fig. 23. MAX-DOAS can be applied by sequentially scanning spectra from different geometries or simultaneous measurements using an imaging spectrometer with 2D-CCD detector coupled to several telescopes. The combination of different elevation angles and two instruments at different locations allows a derivation of the exact plume distance as well. Examples of typical MAX-DOAS scans across a volcanic plume are shown in Fig. 24 (Bobrowski et al., 2003). Trace gas column densities increase strongly when the MAX-DOAS viewing direction crosses the plume, while background column densities are measured outside the plume.

The advantage of MAX-DOAS over traditional passive techniques (i.e. COSPEC, Millan et al., 1969; Davies, 1970) is the much higher time resolution and the reduced effect of fluctuations inside the plume on the measurement results. Additionally many trace gases (such as SO_2 , NO_2 , CH_2O , BrO , CS_2 rather than just SO_2 or – after refitting the instrument with another mask – NO_2) can be measured simultaneously with MAX-DOAS. Recent instrument developments allow the use of rugged, lightweight MAX-DOAS instruments with relatively low power consumption (Galle et al., 2003; Bo-

browski et al., 2003). Compared to active DOAS measurements MAX-DOAS requires much simpler instrumentation, yields profile information, however gives less definite concentration data, and is only useable during daylight hours.

9. Conclusions

5 In this study we have shown that Multi Axis DOAS is an emerging, new variant of the scattered light DOAS method. It is based on mature instrumental techniques which have been established over the last decades albeit for other purposes. Although its development is still ongoing, MAX-DOAS has proven to be successful in several applications including monitoring volcanic plumes (SO_2 , BrO), natural emission of trace
10 gases (e.g. reactive bromine, BrO) from salt lakes, sea ice, and the ocean surface as well as for pollution monitoring. Further applications like study of urban or forest fire plumes, studies of three dimensional trace gas distributions (von Friedeburg, 2003), balloon borne applications, or study of radiative transfer in clouds still remain to be fully explored.

15 Although MAX-DOAS cannot replace active DOAS or other complimentary measurement techniques (e.g. for nighttime observations an active system is needed), in many cases MAX-DOAS is a powerful alternative to traditional methods. As shown in this paper, there are numerous applications where successful measurements have been performed using MAX-DOAS and, in fact, a series of measurements has become only
20 possible because of MAX-DOAS. Particular advantages of MAX-DOAS are the simple and compact instrumentation, the easy deployment and low power consumption (passive technique), the fact that vertical profile information of gases up to several kilometres above the ground can be derived in combination with RTM calculations and the high time resolution also during low visibility conditions. Using advanced radiative
25 transfer models, the slant column densities derived for the various MAX-DOAS viewing directions can be interpreted and combined to yield a more complete data product than other measurement platforms can provide. Additionally, information on aerosol profiles

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and properties can be obtained from MAX-DOAS O₄ (and O₂) measurements which are a by-product of most MAX-DOAS applications. With these aerosol data, necessary input for the RTM can be taken from the very measurement without need for further instrumentation. In the future, enhanced data analysis algorithms for MAX-DOAS combining spectral analysis, radiative transfer modelling and inversion techniques for profile retrieval will further extend the capabilities of MAX-DOAS.

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Table 1. Overview and history of the different scattered light passive DOAS applications, # Axis indicates the number of different viewing directions combined.

Method	Measured quantity	No. of axes, technique	References
COSPEC	NO ₂ , SO ₂ , I ₂	1, (S)	Millan et al. (1969), Davies (1970), Hoff and Millan (1981), Stoiber and Jepsen (1973), Hoff et al. (1992)
Zenith scattered light DOAS	Stratospheric NO ₂ , O ₃ , OCIO, BrO, IO	1	Noxon (1975), Noxon et al. (1979), Harrison (1979), McKenzie and Johnston (1982), Solomon et al. (1987a, b, 1988, 1989, 1993), McKenzie et al. (1991), Fiedler et al. (1993); Pommereau and Piquard (1994), Aliwell et al. (1997); Eisinger et al. (1997); Kreher et al. (1997); Richter et al. (1999); Wittrock et al. (2000)
Zenith sky + Off-Axis DOAS	Stratospheric OCIO	2	Sanders et al. (1993)
Off-Axis DOAS	Stratospheric BrO profile	1	Arpaq et al. (1994)
Zenith scattered light DOAS	Tropospheric IO, BrO	1	Kreher et al. (1997); Friess et al. (2001, 2003), Wittrock et al. (2000)
Off axis DOAS	Tropospheric BrO	1	Miller et al. (1997)
Sunrise Off-Axis DOAS+ direct moonlight	NO ₃ profiles	2, S	Weaver et al. (1996), Smith and Solomon (1990), Smith et al. (1993)

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Table 1. Continued.

Method		Measured quantity	No. of axes, technique	References
Sunrise Off Axis DOAS		Tropospheric NO ₃ profiles	1	Kaiser (1997), von Friedeburg et al. (2002)
Aircraft-DOAS		Tropospheric BrO	2	McElroy et al. (1999)
Aircraft zenith sky + Off-axis DOAS		“near in-situ” stratospheric O ₃	3	Petritoli et al. (2002)
AMAX-DOAS		Trace gas profiles	8+, M	Wagner et al. (2002), Wang et al. (2003), Heue et al. (2003)
Multi DOAS	Axis	Tropospheric BrO profiles	4, S	Hönninger and Platt (2002), Hönninger et al. (2003a)
Multi DOAS	Axis	Trace gas profiles	2–4, M	Löwe et al. (2002), Oetjen (2002), Heckel (2003)
Multi DOAS	Axis	NO ₂ plume	8, M	von Friedeburg (2003)
Multi DOAS	Axis	BrO in the marine boundary layer	6, S/M	Leser et al. (2003), Bossmeyer (2002)
Multi DOAS	Axis	BrO and SO ₂ fluxes from volcanoes	10, S	Bobrowski et al. (2003)
Multi DOAS	Axis	BrO emissions from Salt Lake	4, S	Hönninger et al. (2003b)

S = Scan, M = Multiple telescopes

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Table 2. Summary of vertical profile shapes used in this study.

Profile number	Profile shape	Atmospheric counterpart
P1	$c = c_0$ for 0–1 km, $c = 0$ above	Absorber confined to well mixed 1 km boundary layer BL
P2	$c = c_0$ for 0–2 km, $c = 0$ above	Absorber confined to well mixed 2 km BL
P3	$c = 2c_0$ for 0–1 km, $c = c_0$ for 1–2 km, $c = 0$ above	Absorber mixing from BL to higher altitudes
P4	$c = 0$ for 0–1 km, $c = c_0$ for 1–2 km, $c = 0$ above	Absorber present in layer lifted from the ground
P5	$c \propto [c(\text{O}_2)]^2$	O_4 profile
P6	Gaussian shape around 25 km altitude, 10 km FWHM	Stratospheric absorber (e.g. O_3 , NO_2)
P7	$c = 0$ for 0–3 km, $c = c_0$ for 3–5 km, $c = 0$ above	Absorber in lower free troposphere
P8	$c = 0$ for 0–3 km, $c = c_0$ for 7–9 km, $c = 0$ above	Absorber in higher free troposphere

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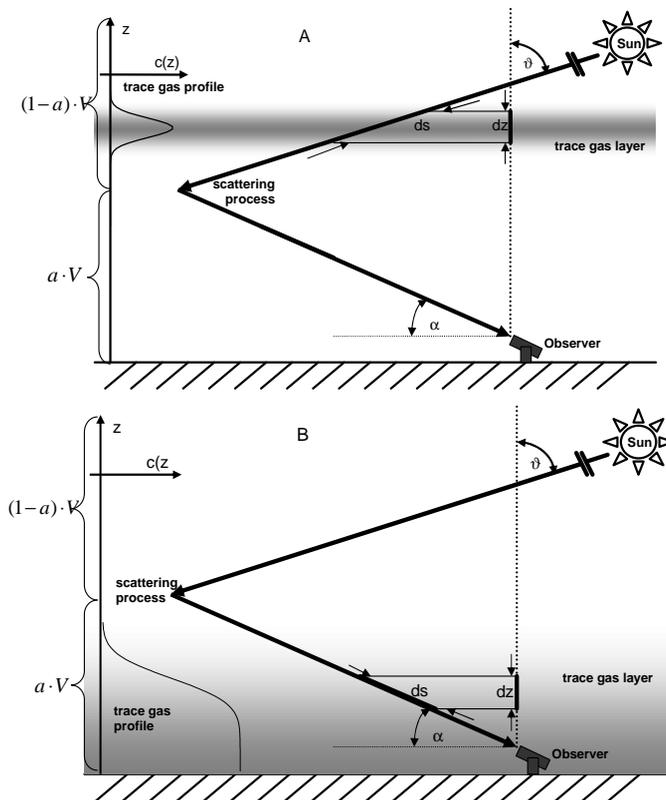


Fig. 1. Observation geometries for ground based DOAS using scattered sunlight: Light enters the atmosphere at a certain solar zenith angle ϑ . In the single scattering approximation light received by the observer was scattered exactly once into the telescope viewing direction defined by the observation elevation angle α . The observed SCD (integral along ds) is larger than the VCD (integral along d_z), with AMF being the conversion factor. Panel A represents the situation for a high (stratospheric) trace gas layer, panel B is representative for a trace gas layer near the surface.

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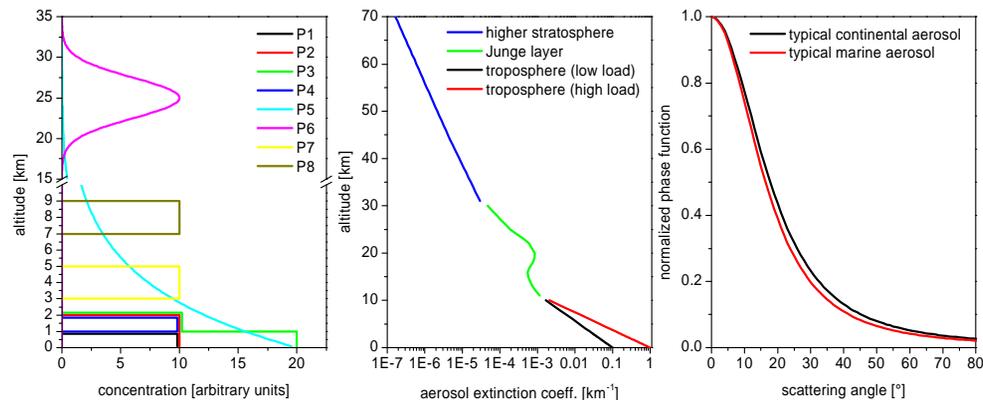


Fig. 2. Profile shapes of test profiles P1–P8 (left), aerosol profile shapes (centre) and aerosol scattering phase functions (right) used for this study (see text).

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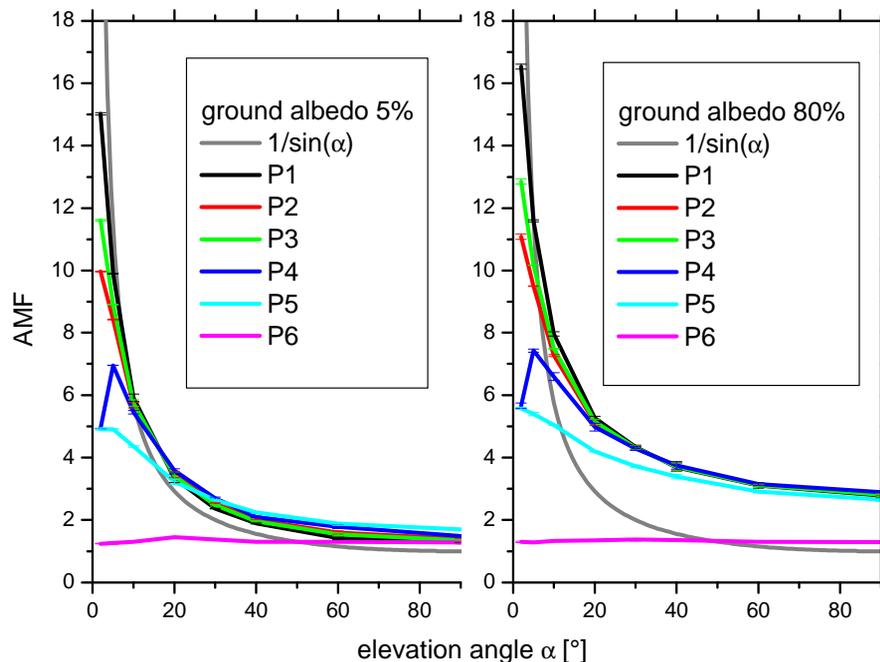


Fig. 3. AMF dependence on the viewing direction (elevation angle α) for the profiles P1–P6 (see Table 2) for 5% ground albedo (left) and 80% albedo (right), respectively. Calculations were performed for a wavelength of 352 nm, 30° Solar zenith angle, and assuming a pure Rayleigh case for the troposphere. For the stratosphere (i.e. above 10 km) the standard aerosol scenario (Fig. 2, centre panel) was assumed. The error is the intensity weighted standard deviation of the AMF averaged over the modelled photon paths. The $1/\sin(\alpha)$ approximation is indicated for reference.

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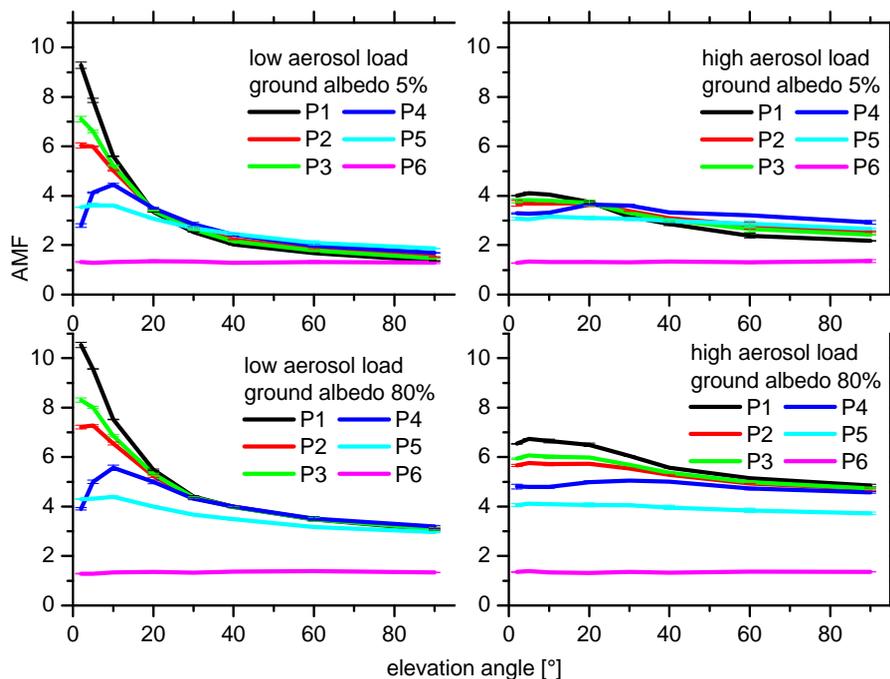


Fig. 4. AMF as a function of viewing direction (elevation angle α) for P1–P6 for low (left column) and high (right column) aerosol load and for 5% (top panels) and 80% (bottom panels) ground albedo, respectively. Calculations were performed for the same scenario as in Fig. 3, but for typical continental aerosol (left panels: low aerosol load, right panels: high aerosol load, see Fig. 2), differences to marine aerosol are discussed below.

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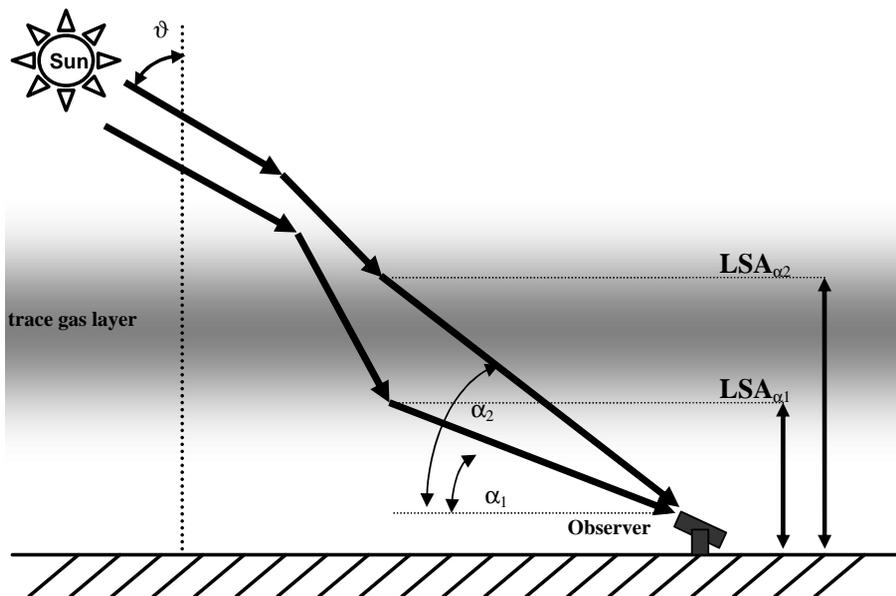


Fig. 5. The Last Scattering Altitude LSA: for low elevation angles, the mean free path in the viewing direction is shorter due to higher density and/or aerosol load. This can result in the slant path through absorbing layers at higher altitudes being shorter for lower elevation angles than for higher ones.

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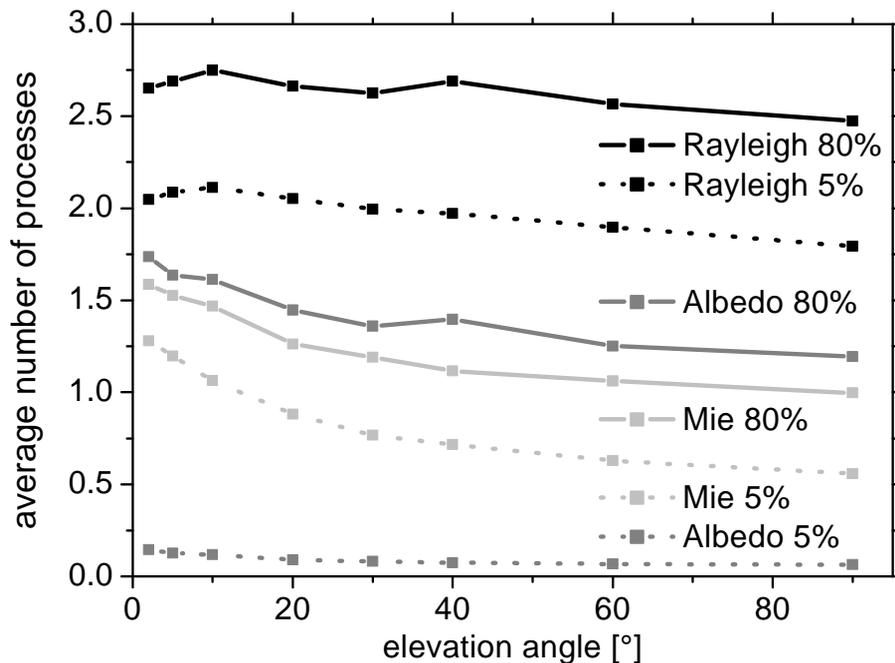


Fig. 6. Number of Rayleigh, ground (albedo), and Mie scattering events as a function of elevation angle for the typical rural low aerosol scenario and 5% (dashed lines) and 80% (drawn lines) ground albedo, respectively.

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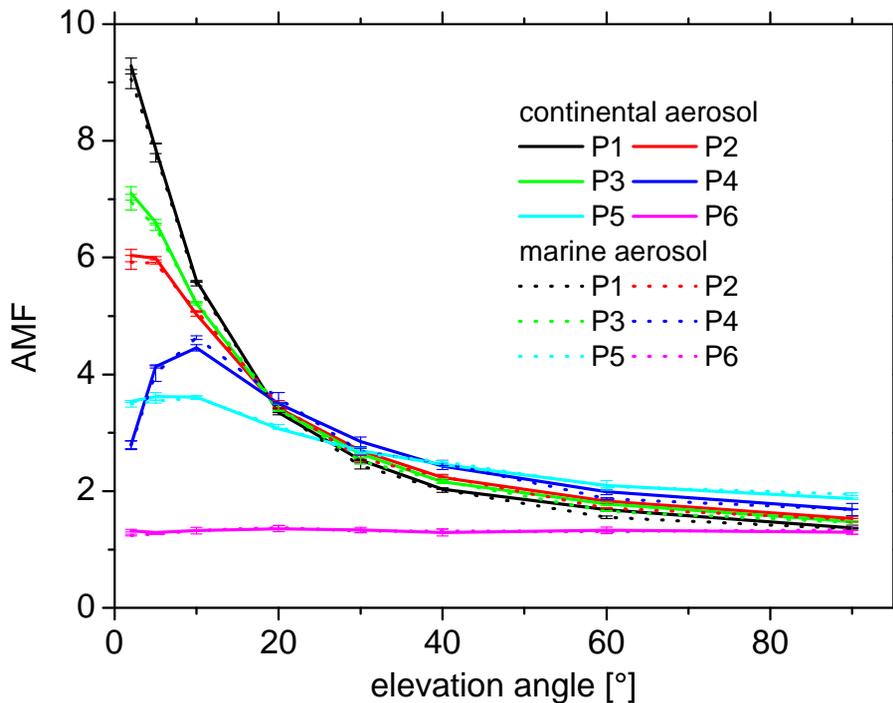


Fig. 7. Effect of different aerosol types on the AMF, drawn lines indicate continental background aerosol (same as “low aerosol load” in Fig. 4). Only small differences result from different scattering phase functions.

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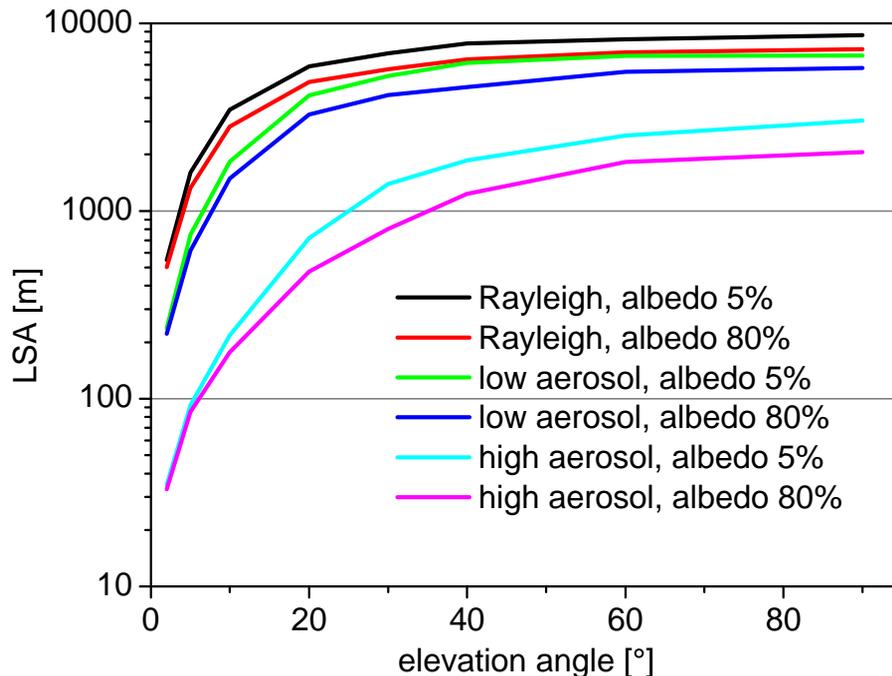


Fig. 8. Last scattering altitude (LSA) for pure Rayleigh, low and high aerosol load scenarios (see Fig. 4) and both, 5% and 80% albedo. The LSA is generally below 1km for the lowest elevation angle and above 1km for the highest elevation angles, especially for zenith viewing direction.

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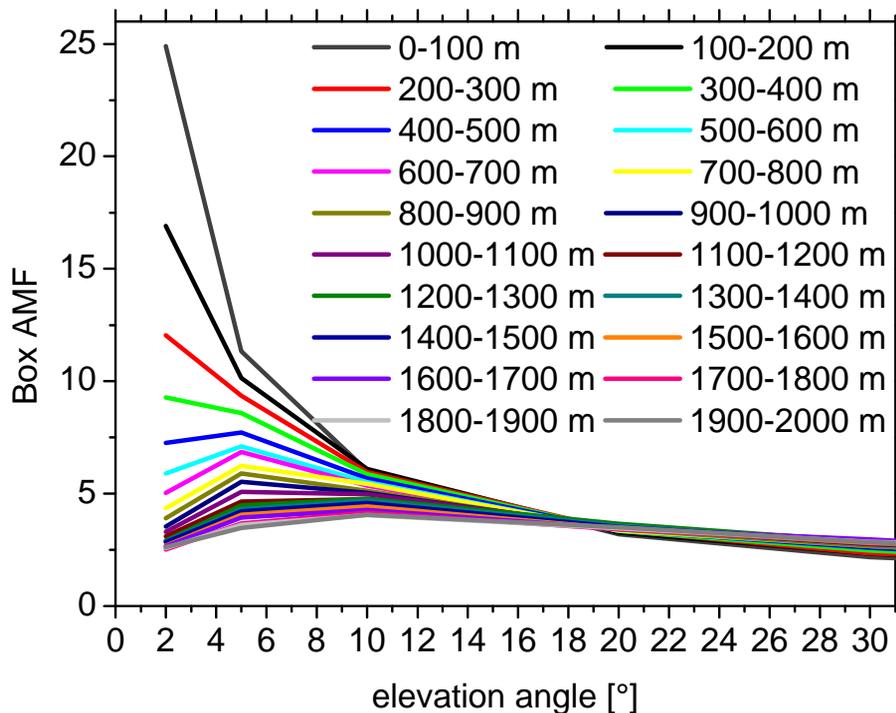


Fig. 9. Box AMF for the model layers below 2 km altitude. The sensitivity towards the lowest layers changes strongly for elevation angles below 10°, for elevation angles smaller than 5° the sensitivity already decreases for layers above 400 m altitude.

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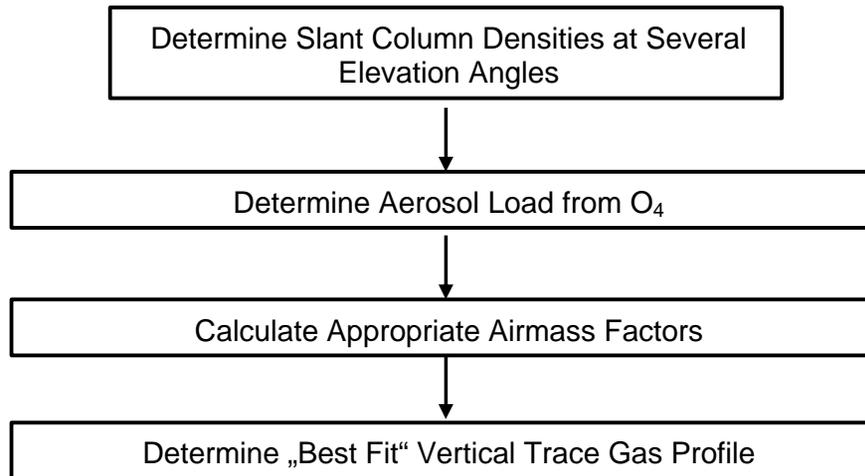


Fig. 10. Diagram of a MAX-DOAS evaluation procedure including the determination of the aerosol load from O₄ observations.

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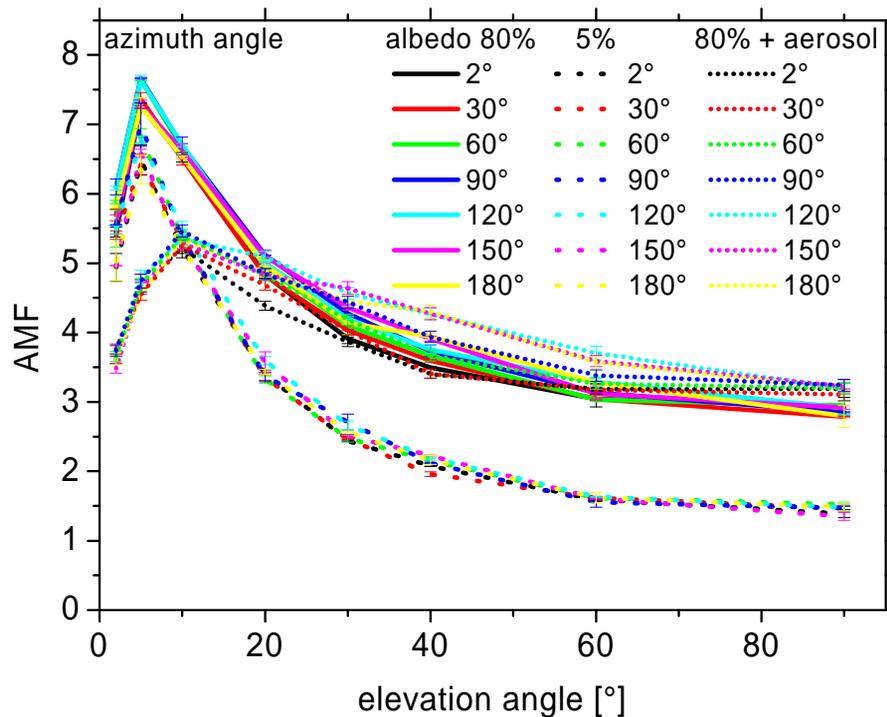


Fig. 11. Azimuth-dependence of the AMF for 30° SZA and profile P4 (trace gas layer at 1–2 km altitude). For relative azimuth angles between 2° (looking almost towards the sun) and 180° (looking away from the sun) only a small effect increasing with ground albedo and aerosol load can be seen. Calculations were performed for a ground albedo of 80% and 5% (cf. Fig. 3) in the Rayleigh case (in the troposphere) and for 80% ground albedo in the low aerosol case (cf. Fig. 4).

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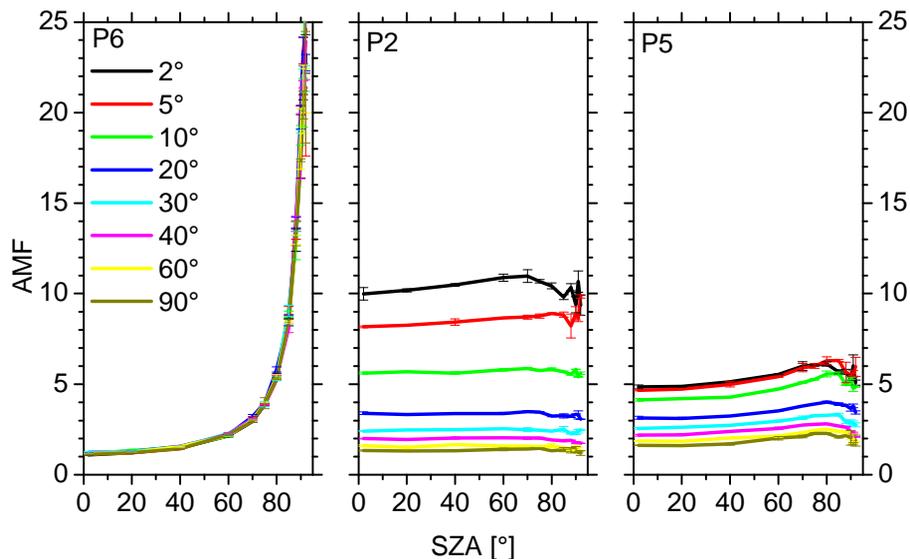


Fig. 12. Solar Zenith angle (SZA) – dependence of the AMF for the typical stratospheric profile P6 and the boundary layer profile P2 as well as the O_4 profile P5 for comparison. The expected strong dependence is observed for the stratospheric absorber, with no significant dependence on the viewing direction. In contrast to that for the tropospheric profiles P2 and P5 significant differences for the various viewing directions can be seen, whereas the SZA dependence is significant only at higher SZA (i.e. $> 70^\circ$).

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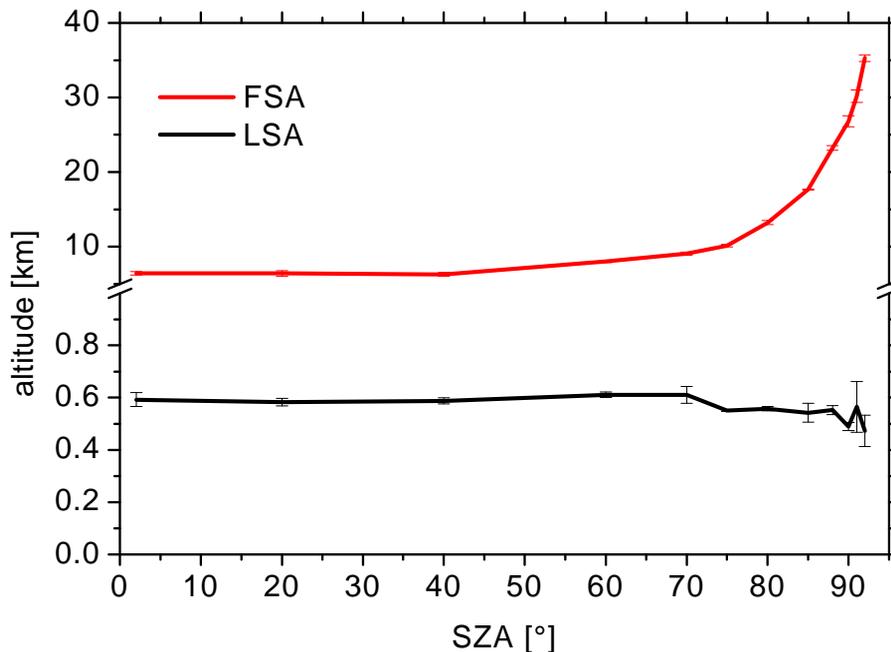


Fig. 13. First and last scattering altitude for the 2° elevation angle. The FSA strongly depends on the SZA, whereas the LSA is largely independent of the SZA. Note the axis break at 1km altitude and the expanded y-scale below.

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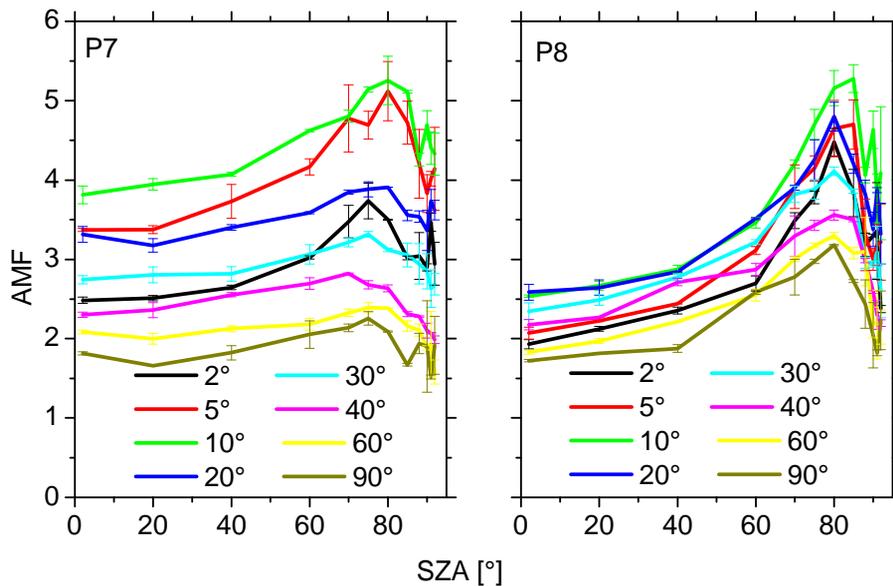


Fig. 14. AMF as a function of SZA for the “free tropospheric profiles” P7 and P8 with the elevation angle as parameter (shown in different colors). The SZA dependence is smaller than for the stratospheric profile P6, but much stronger than for the lower tropospheric profiles P1–P5.

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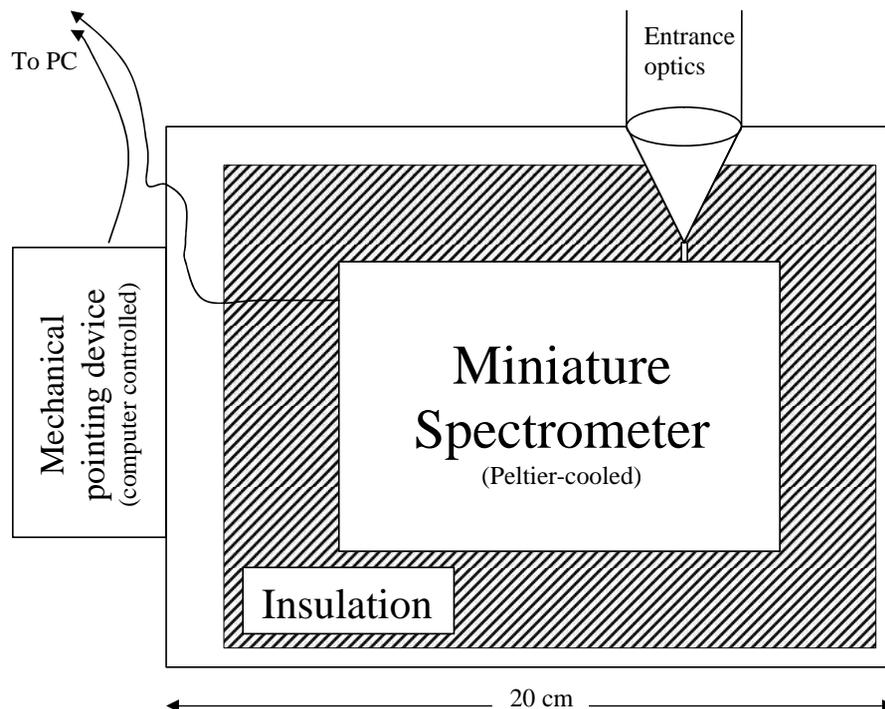


Fig. 15. Outline of the Mini-MAX-DOAS instrument. A miniature spectrometer is cooled and temperature stabilized by a 2 stage Peltier cooler. The built-in entrance optics (telescope) provide a narrow field of view ($< 0.5^\circ$). The unit can be pointed in various directions by computer control.

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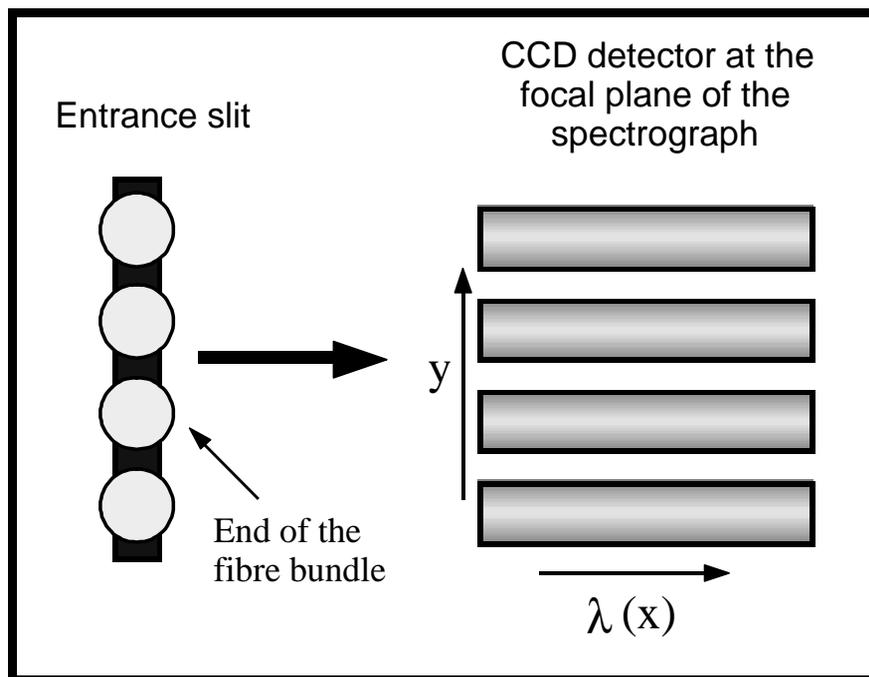


Fig. 16. Two dimensional spectrometer: Orientation of the spectra of the measured light from different viewing angles on the CCD array. The dispersion direction is labeled “x”, thus for each fibre ending at the entrance slit a complete and separate spectrum is generated on the CCD.

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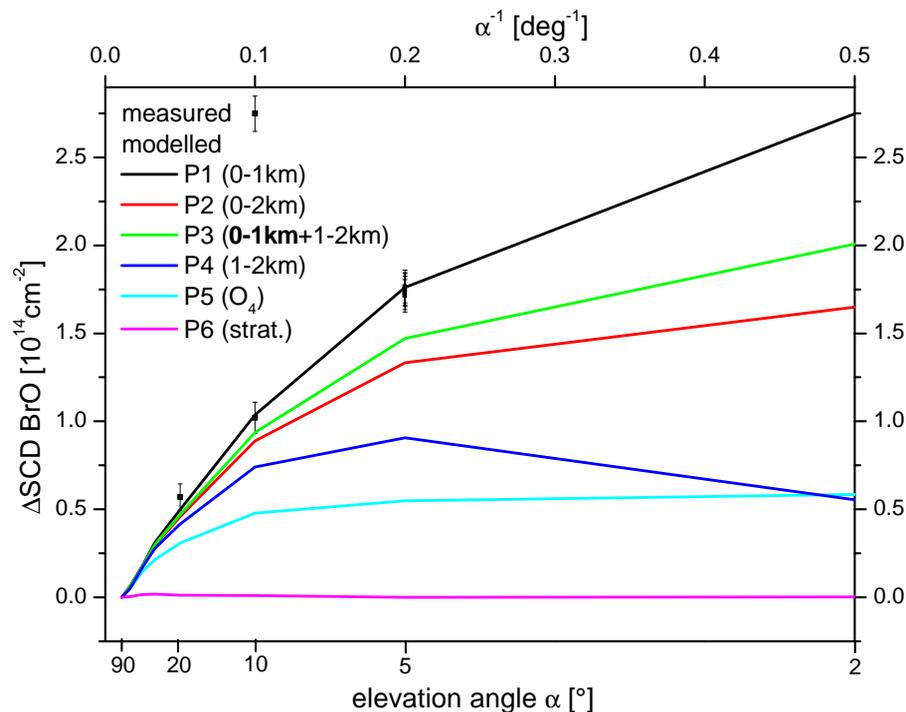


Fig. 17. Comparison of ΔSCD 's measured on 4 May 2000 (15:15 UT–15:40 UT) during the Alert2000 field experiment by Hönninger and Platt (2000) with calculated ΔSCD 's for assumed profile shapes P1–P6 as a function of the elevation angle α . For better illustration the x-axis was chosen linear in α^{-1} . 4 May 2000 at Alert was characterized by a relatively constant BrO column ($\text{BL-VCD}_{\text{BrO}} = 2 \times 10^{13} \text{ molec/cm}^2$). Only P1 is compatible with the measured values.

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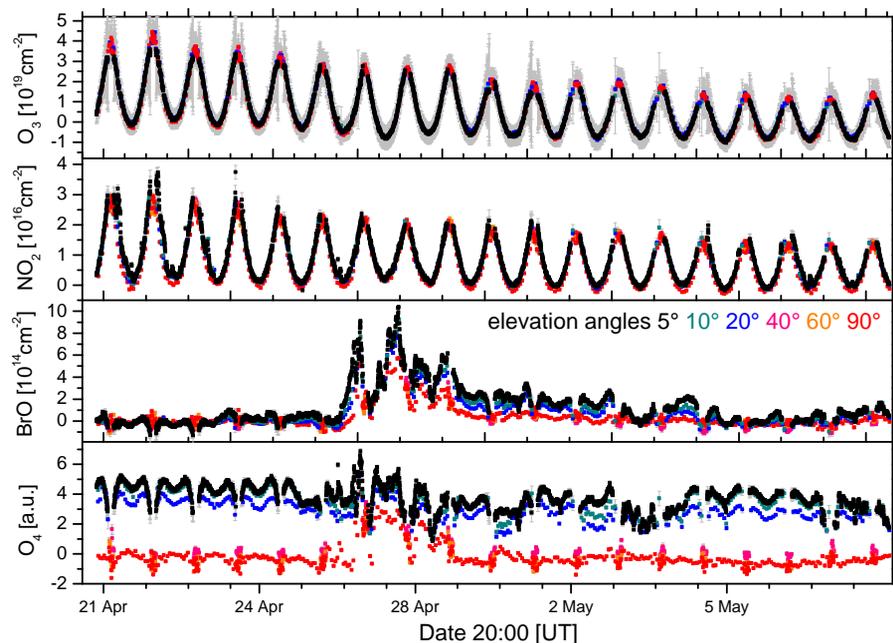


Fig. 18. MAX-DOAS time series of DSCD's of O_3 , NO_2 , O_4 and BrO at Alert during spring of 2000. The O_3 and NO_2 columns show no variation for the different elevation angles indicating that the bulk of these species is located in the stratosphere. In contrast to that the O_4 data vary strongly with elevation angle. Enhanced O_4 levels around 27/28 April at high elevation angles indicate periods of snowdrift. Following 27 April the BrO columns show strong differences for different elevation angles due to BrO presence in the boundary layer. Effects of snowdrift on 27/28 April can be seen like for O_4 .

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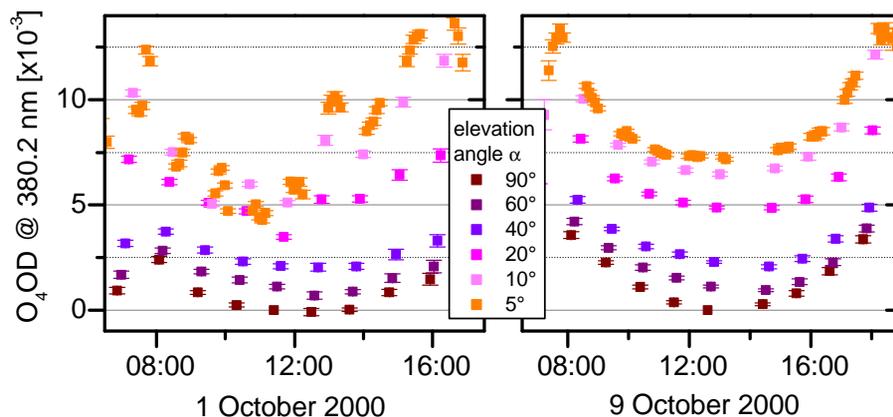


Fig. 19. O_4 absorption at different elevation angles in mid-latitudes. Clearly visible is the pattern of increasing O_4 absorption with decreasing elevation angle. The regular pattern shown for a cloud-free day on the right is disturbed only for the lowest elevation angles on a cloudy day (left). The Multi Axis effect is much stronger than the overlaid U-shape pattern which is due to the change in SZA (Leser, 2001; Leser et al., 2003).

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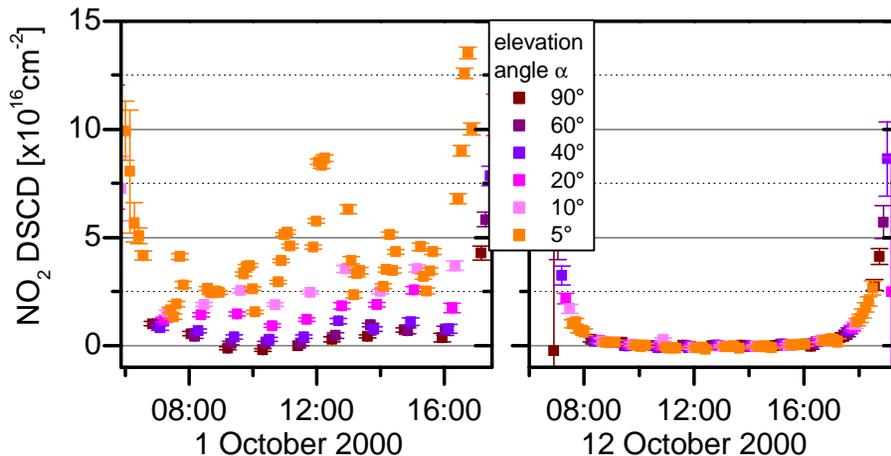


Fig. 20. Shown are two examples of NO_2 DSCD's, one for a case with pollution observed in the lower troposphere (left panel, North Sea, 1 October 2000), the right panel shows stratospheric background NO_2 only with no significant differences between the individual elevation angles (Leser, 2001; Leser et al., 2003).

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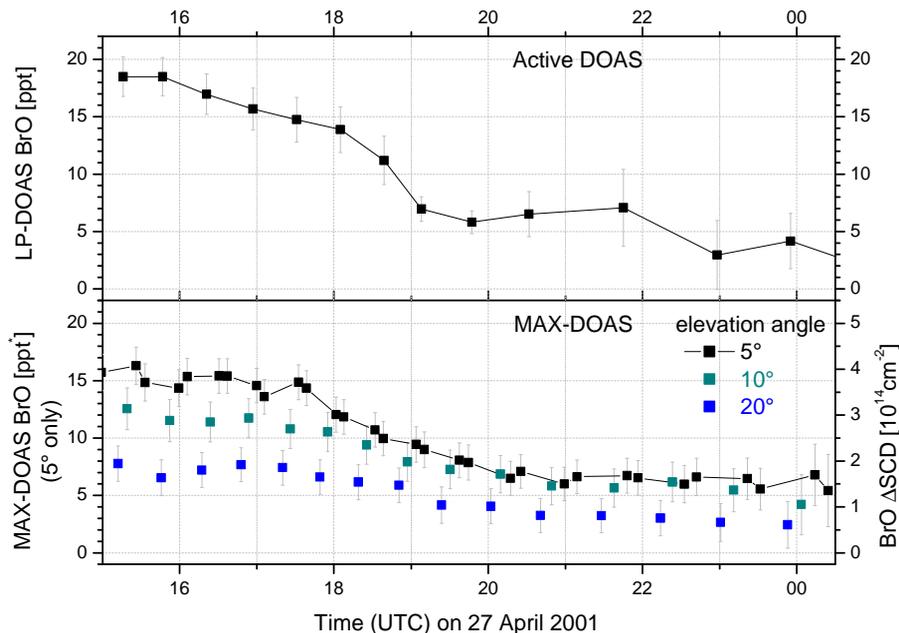


Fig. 21. Comparison of active long path DOAS and passive MAX-DOAS BrO measurements at the Hudson Bay for 27 April 2001. From the elevation angle dependence of the ΔSCD 's a ~ 1 km BrO layer at the surface can be concluded for e.g. 16:00 h. Using this layer thickness the 5° BrO ΔSCD 's were converted to mixing ratios for better comparison with the long path DOAS data (Hönninger et al., 2003a).

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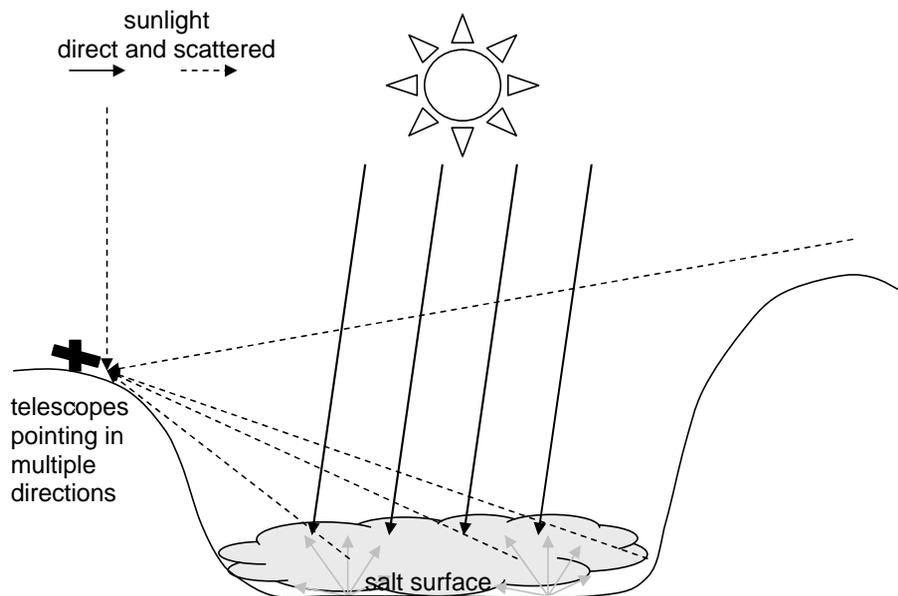


Fig. 22. Salt flat MAX-DOAS set-up: Horizontal and vertical emission patterns of halogen compounds can be measured.

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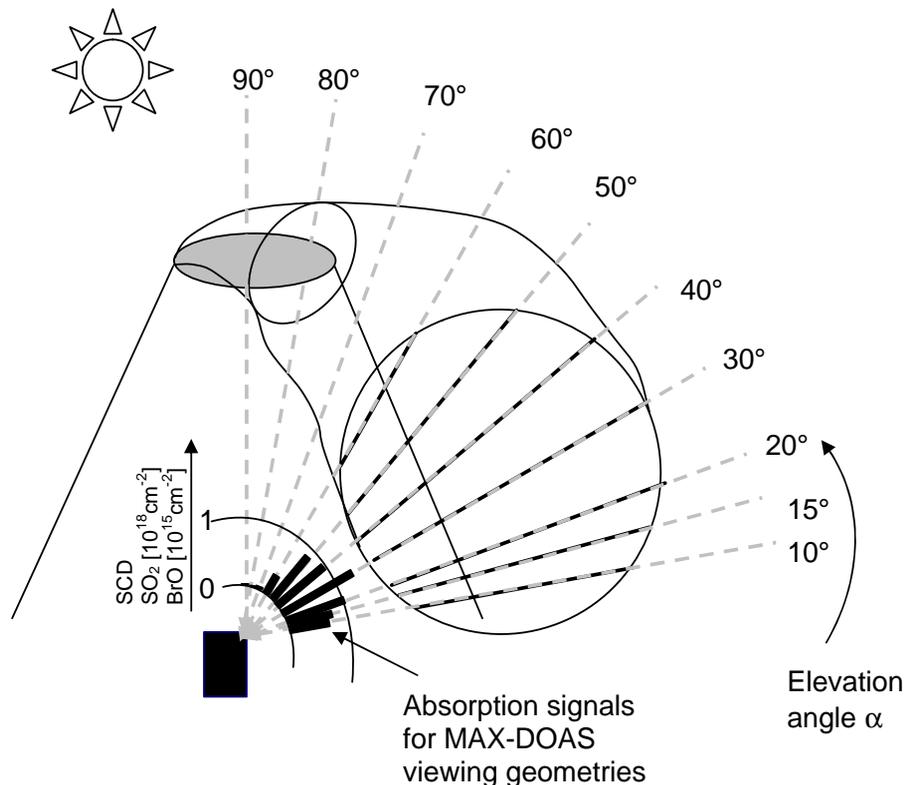


Fig. 23. Emission plume monitoring by MAX-DOAS: A volcano as emission source is shown as an example. MAX-DOAS measurements yield spatial information as well as concentration and emission flux data for several trace gases. As an example the viewing directions used by (Bobrowski, 2002; Bobrowski et al., 2003) are sketched and the distribution of the SCD signals of the first scan of Fig. 24 for BrO is included.

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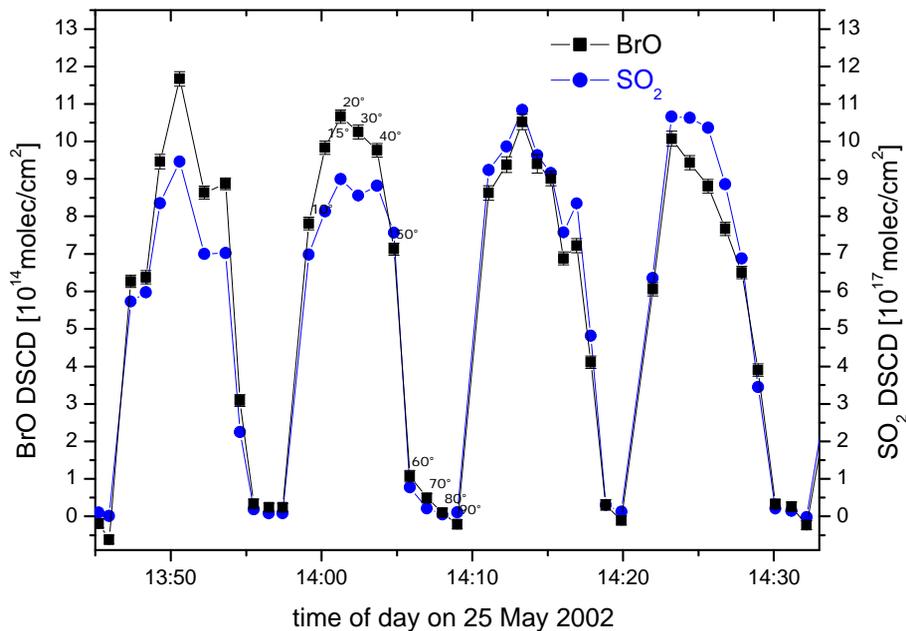


Fig. 24. Example of a series of MAX-DOAS scans across a volcanic plume. Volcanic gases like SO_2 and BrO show high DSCD's when the viewing direction crosses the plume, background values are found outside the plume (Bobrowski, 2002; Bobrowski et al., 2003). For the second scan the BrO data points are labelled with the respective MAX-DOAS elevation angle.

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