tion of the received light is reflected by the target, or, alternatively, black targets can be used; then the received light is scattered solely within the air volume between the instrument and target. In either case, the novel technique combines the advantages of both passive and active systems; no artificial light sources involving relatively complicated optical setups are needed. In addition, the interpretation of the observations can be referred to well-defined light paths and even allows tomographic inversions.[26]

The main limitations of the method are the restrictions to wavelengths >300 nm and to daytime. In particular, most aromatic compounds and nighttime chemistry cannot be investigated. However, there is a large class of species including NO_2, HCHO, SO_2, H_2O, Glyoxal, BrO, and others that can be detected.

The paper is organized as follows: In Section 2 we introduce the proposed measurement technique, describe the composition and information content of the measured light, and the specific requirements and subsequent steps of the spectral analysis. In Section 3 the various measurements strategies and possible results (including tomographic techniques) are described. In Section 4 the first measurement examples are presented. Finally, in Section 5 we summarize our findings and give recommendations for basic and advanced measurement strategies and for the specific design of advantageous experimental setups.

2. Multiaxis Differential Optical Absorption Spectroscopy Observations of Illuminated Targets

In many aspects tomographic MAX-DOAS observations can be treated very similarly to tomographic DOAS measurements using artificial light sources and well-defined light paths.[1–3,27] In particular, we will show in this paper that tomographic MAX-DOAS eventually yields an intermediate product very similar to active long-path DOAS: the path-integrated (or average) trace gas concentration between the instrument and the target. However, there are also two fundamental differences: First, MAX-DOAS observations use the Sun, which is an extraterrestrial light source; second, atmospheric scattering processes between the instrument and the target can also contribute to the measured signal. In the following subsections the resulting effects and their correction are described in detail.

A. Tomographic Multiaxis Differential Optical Absorption Spectroscopy Measurements

MAX-DOAS observations directed to targets illuminated by sunlight receive photons that have traversed two basic sections in the atmosphere: the distance between the top of the atmosphere and the target, and the distance between the target and the instrument (see Fig. 1).

Depending on the investigated problem, the properties of the measuring site, and the number of instruments and targets, a large variety of measurement geometries can be developed. In the simplest case, one instrument is directed to one target; then the average concentration between the instrument and the target can be derived. Targets at different distances or altitudes allow the retrieval of horizontal or vertical gradients (see Fig. 1). Higher-dimensional tomographic setups can even yield the spatial trace gas distribution.

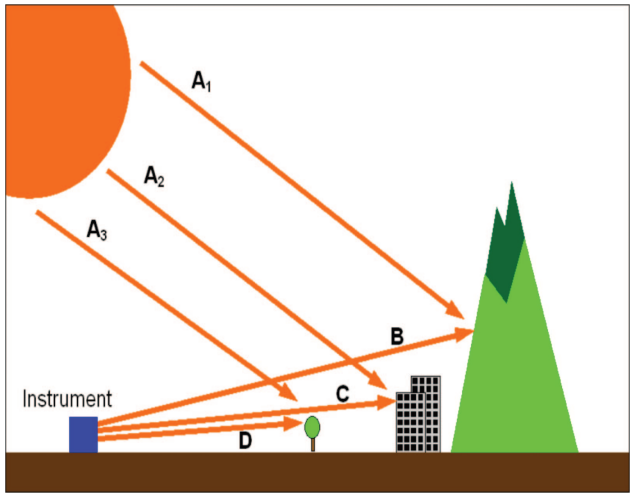

Fig. 1. (Color online) Experimental setup for tomographic MAX-DOAS observations. In contrast to conventional MAX-DOAS observations the telescope is directed toward targets that are illuminated by the Sun. The total signal contains the trace gas absorptions of two sections of the total light path: between the targets and the top of the atmosphere (paths A_1, A_2, A_3) and between the instrument and the targets (paths B, C, D). After correction for the absorption from the section between the targets and the top of the atmosphere (see Subsection 2.B) the average trace gas concentration between the instrument and the target can be analyzed. If several targets are used, even multidimensional trace gas distributions can be retrieved.

Irrespective of the final inversion strategy, the basic quantity that is derived from the spectral DOAS analysis is the (differential) optical depth τ of a selected trace gas, from which the so-called slant column density (SCD) S, the trace gas concentration integrated along the light path L, can be derived by dividing it by the respective absorption cross section at the same wavelength:

$$S = \int_0^L \rho(l)\mathrm{d}l = \frac{\tau(\lambda)}{\sigma(\lambda)}. \quad (1)$$

It should be noted that for the DOAS analysis the so-called differential optical depth is usually analyzed, which, in the simplest case, is the difference of the optical depth at different wavelengths.[14,28] In analogy, a differential cross section can be defined, which then has to be applied in Eq. (1). It might, for example, indicate the difference of the absorption cross section (or optical depth) inside and outside an absorption band. For these differential quantities we will hereafter use the terms τ' and σ'.

The analyzed total SCD can be expressed as the sum of the partial SCDs of both sections introduced above (see Fig. 1):

$$S_{\mathrm{tot}} = S_{\mathrm{Target}} + S_{\mathrm{Atm}}. \quad (2)$$

Here S_{Target} is the trace gas concentration integrated between the instrument and the target, and S_{Atm} is the trace gas concentration integrated between the target and top of the atmosphere. For many trace gases such as NO_2, BrO, or O_3, a substantial or major fraction of the total atmospheric column is located in the stratosphere; for such trace gases, S_{Atm} will be independent from the MAX-DOAS viewing direction because the absorption paths through the stratosphere are determined by the solar zenith angle. Even for trace gases located in the free or upper troposphere this assumption is valid with only small restrictions.

It should be noted that the sensitivity of the measurement for the trace gases between the instrument and the target strongly depends on the brightness of the target and the atmospheric conditions. Only for white targets is the measured SCD almost similar to the true SCD S_{Target} (see Subsection 2.C.1).

Since we are interested in S_{Target}, we have to remove S_{Atm} from the total signal. This can be easily achieved and will be explained in Subsection 2.B. Another fundamental question is to determine the sensitivity of the measurement for the trace gas between the instrument and the target, in particular the dependence of the sensitivity as a function of the distance from the instrument. For an ideal measurement the sensitivity would be constant along the light path between the instrument and the target as, for example, for traditional long-path DOAS observations. We will show below that for many cases the sensitivity can be expressed as a simple functional expression of the distance from the instrument. For bright targets, for example, the sensitivity becomes similar to that for long-path DOAS observations. Nevertheless, in general, this sensitivity decreases with the distance from the detector because additional sunlight is scattered into the line of sight of the instrument (see Fig. 1). For the correct interpretation of the measurement, this sensitivity has to be characterized (see Subsection 2.C) and corrected. If no simple correction is possible, radiative transfer modeling has to be applied (see Subsection 2.D).

B. Removal of the Signal From the Region Between the Target and Top of the Atmosphere (S_{Atm})

The correction of the partial SCD between the target and the top of the atmosphere (S_{Atm}) can be easily (and quasi automatically) achieved by two procedures. The main aspect of both methods is that for the DOAS analysis of scattered sunlight, a so-called Fraunhofer spectrum has to be included for the correction of the strong solar Fraunhofer lines.[8,9,11,23] For this purpose, usually a measurement taken with the same instrument but under different measurement conditions (e.g., at a low solar zenith angle) is used. Since this Fraunhofer spectrum contains not only the solar Fraunhofer lines but also atmospheric absorptions, the result of the spectral DOAS analysis is the difference between the SCDs of the measurement and the Fraunhofer spectrum. The trick for the correction of signal from between the target and the top of the atmosphere (S_{Atm}) is to select a Fraunhofer spectrum that contains only S_{Atm}. This can easily be achieved by two simple approaches. The first approach is to point the telescope of the instrument toward the zenith. It then receives photons that have traversed almost similar paths as those between the target and the top of the atmosphere. Especially for trace gases that (in addition to the boundary layer) are located only in the stratosphere, a zenith-scattered spectrum is already sufficient for the correction of S_{Atm}. If trace gases are also located in the free troposphere it might be better, however, to choose a refined approach. For the measurement of the Fraunhofer spectrum the telescope should then be directed to a target close to the instrument, which has similar properties compared to the real targets. Particularly the orientation (the angle of the reflecting surface with respect to the instrument and the Sun) and the reflectivity (the albedo) of the nearby target should be similar to the target at far distance. This procedure ensures that the photon paths in the lower free troposphere (and thus the S_{Atm}) of both the actual measurement and the Fraunhofer spectrum are almost identical.

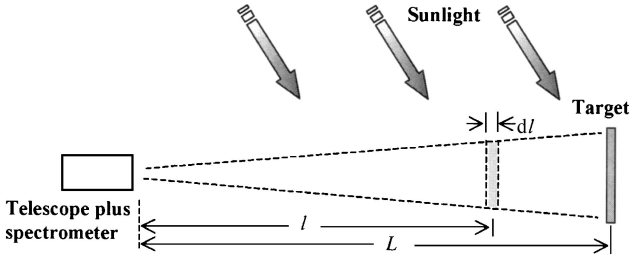

Fig. 2. Composition of the light received by the telescope: sunlight can be reflected by the target or scattered by molecules and aerosols between the target and the instrument.

In some cases it might be sufficient to use a measurement from one (far) target (e.g., measurement D in Fig. 1) as the Fraunhofer spectrum for the analysis of a measurement from another (far) target that is placed in the same direction but at a different distance (e.g., measurement C in Fig. 1). Then the resulting S_{Target} is the integrated trace gas concentration along the path between both targets.

C. Sensitivity of the Measurement for Trace Gases Between the Instrument and the Target

For the correct interpretation of the trace gas absorption determined by the DOAS analysis the sensitivity of the MAX-DOAS observation for the trace gas between the instrument and the target (e.g., as a function of the distance from the instrument) has to be known. We show in Subsections 2.C.1–2.C.3 that for many cases simple functional expressions for this dependence can be found. The light received by the telescope $I(\lambda, L)$ contains two parts (see Fig. 2):

$$I(\lambda, L) = I_{\mathrm{Atmos}}(\lambda, L) + I_{\mathrm{Target}}(\lambda, L), \qquad (3)$$

where $I_{\mathrm{Target}}(\lambda, L)$ is the measured radiance caused by the reflected sunlight by the target, and $I_{\mathrm{Atmos}}(\lambda, L)$ is