

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.<sup>26</sup>

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  $\text{NO}_2$ ,  $\text{HCHO}$ ,  $\text{SO}_2$ ,  $\text{H}_2\text{O}$ , Glyoxal,  $\text{BrO}$ , 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.<sup>1–3,27</sup> 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 dif-

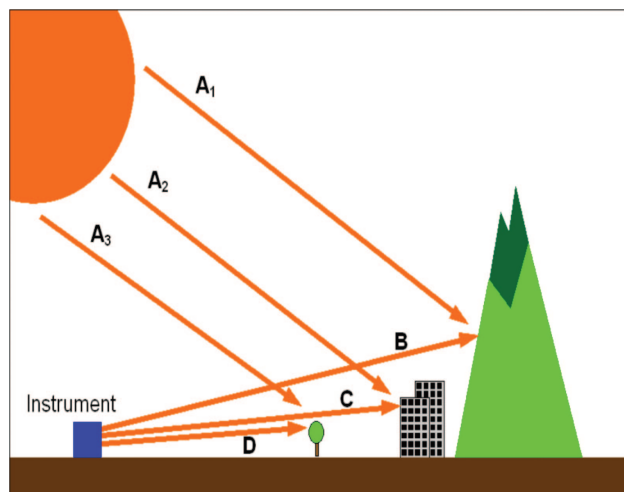


Fig. 1. (Color online) Experimental setup for tomographic MAX-DOAS observations. In contrast to conventional MAX-DOAS observations the telescope is directed toward targets that are illuminated by the Sun. The total signal contains the trace gas absorptions of two sections of the total light path: between 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.

ferent 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.

Irrespective of the final inversion strategy, the basic quantity that is derived from the spectral DOAS analysis is the (differential) optical depth  $\tau$  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:

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

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.<sup>14,28</sup> 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  $\tau'$  and  $\sigma'$ .

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

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