Simultaneous UV-vis Measurements of BrO from Balloon, Satellite and Ground: Implications for Tropospheric BrO

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Abstract

In this study balloon-borne SAOZ-BrO profile measurements have been compared with simultaneous satellite (GOME) and ground-based measurements, performed in the framework of the THESEO project "Stratospheric BrO". A 1D model, initialised using the 3D Chemical Transport Model Slimcat, is used to determine and thereby account for the photochemical induced changes of BrO amount during twilight. Three different approaches have used to investigate the BrO abundances in the free troposphere: 1. Comparison of vertical column densities (VCD's) from balloon and satellite observations, 2. Comparison of VCD's from balloon, satellite and groundbased instruments for selected days with respect to cloudiness, and 3. Comparison of slant column densities (SCD's) from balloon and model.

1. Introduction

Recently it has been suggested that BrO could be present in significant amounts not only in the planetary boundary layer but also in the free troposphere. UV-Vis measurements suggest free tropospheric BrO abundances ranging from background amounts of 1-2 pptv [1 ,2] up to 10 pptv [3] during BrO events. The presence of such BrO concentrations throughout the troposphere has fundamental implications for the tropospheric chemistry. It also implies that BrO total columns derived from ground- or satellite-based measurements comprise weighted stratospheric and tropospheric BrO column components.

2. Measurements

UV-Vis sunlight measurements of BrO have been performed simultaneously from three different platforms (table 1). A similar analysis procedure has been applied to the spectra from the different measurements [4]. For the satellite (GOME) measurement ([5] and references therein), data from pixels inside a cycle of 500 km radius around the balloon launch site are taken into account and averaged. The slant column densities (SCD's) of the groundbased and GOME measurements are converted into vertical column densities (VCD's) using Airmassfactors (AMF's) calculated assuming balloon profiles.

	SAOZ-BrO Balloon	GOME Satellite	Ground-based (GB)
Reference	[6]	[5],[7],[8]	[8], [1]
Viewing geometry	Direct sun	nadir	zenith
Time	Evening	noon	continuously daytime
Result	vertical profiles	total columns	total columns

Table 1. Description of the different platforms.

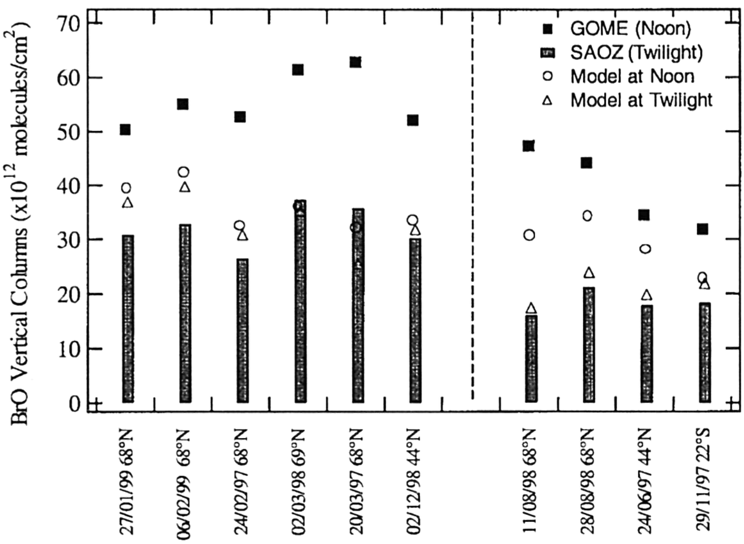

Figure 1. BrO integrated stratospheric profile from SAOZ-balloon measurements (bars) compared to GOME measurements and stratospheric Slimcat model simulations.

3. Results

For the comparison of the measurements performed at different solar zenith angle (SZA), stratospheric 1D photochemical model simulations have been performed (described in [6]).

(i) SAOZ / GOME / MODEL : Vertical columns

When comparing integrated balloon profiles (measured at sunset) with total columns observed by GOME (at noon), systematically higher values of 1-3 $*10^{13}$ molecule/cm^2 are observed for the GOME measurement (Figure 1). The model simulations show that the photochemical decrease can explain only part of the difference (Figure 1). The stratospheric columns, derived from the SAOZ balloon measurements, are in good agreement with the stratospheric model columns as expected from the profile comparison [6]. The differential SCDs from ground-based measurements yield a BrO decrease at sunset, which is well captured by the model [9] (see also Figure 4, right). Thus the differences between GOME and the model are considered to be real. Attributing these differences to tropospheric BrO, yields a tropospheric column of 1-3 $*10^{13}$ molecule/cm^2, which corresponds to 1-2 pptv BrO homogeneously distributed throughout the troposphere.

(ii) SAOZ / GOME / GROUND-BASED : Vertical columns

The determination of the BrO VCD from ground based measurements is subject to relatively large errors [e.g. 7,8,9]. Analyses mostly utilise "differential slant columns", which represent the differences between the columns measured at 80° and 90° SZA [e.g. 9]. In this study the relative changes of the BrO VCD and changes in the cloud cover have been investigated. Figure 2 displays a time series of VCD's measured from ground and satellite over Kiruna [8]. For clear sky conditions the GB columns are relatively high, whereas during cloudy sky conditions they are smaller than those from GOME measurements. Measurements from the three platforms over Kiruna are displayed in figure 3: two DOAS-balloon flights are added [2]. One SAOZ flight was performed over Andoya, however simulations suggest negligible differences between the two launch sites for the stratosphere. The GB measurements yield relatively low VCD's during five cloudy days and relatively high VCD's during the two clear sky days, implying a significant change of the BrO column in the presence or absence of clouds.

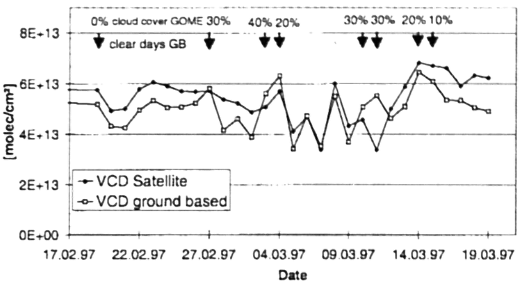

Figure 2. Time series of ground-based (GB) and GOME measurements over Kiruna

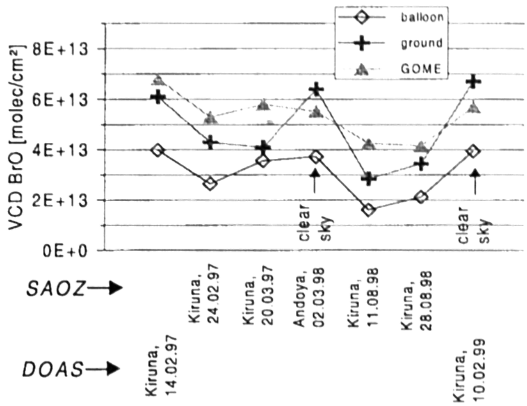

Figure 3. Vertical columns measured from ground, balloon and satellite over Kiruna/Andoya.

In Figure 4 the diurnal variation of the vertical column observed from the ground is compared to the simulation for a cloud-free day. The BrO derived from GOME and SAOZ observations are included. The left panel displays the total vertical columns assuming a tropospheric background of 0 pptv BrO; the right panel shows those assuming a tropospheric background of 3 pptv BrO. Agreement between all measurements can only be achieved, if BrO is assumed to be present in the troposphere.

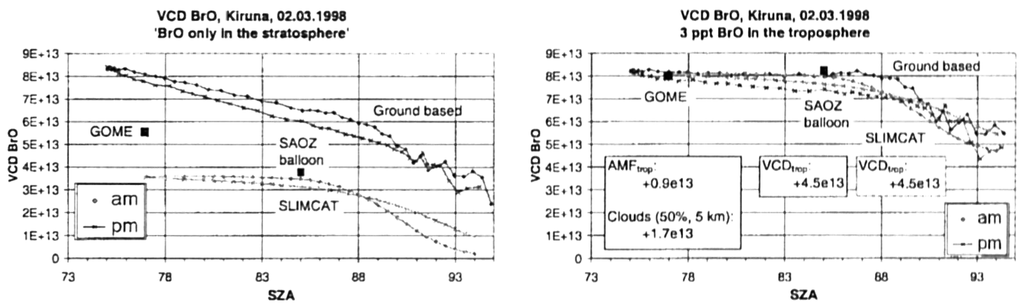

Figure 4. Diurnal variation of the vertical BrO column over Kiruna on a clear day (02/03/98): Groundbased, GOME-satellite and SAOZ-balloon measurements and SLIMCAT model simulation. Left: Results assuming only stratospheric BrO. Right: Results assuming tropospheric BrO (with constant mixing ratio of 3 pptv).

(iii) SAOZ /MODEL : Slant columns

The model output was used to calculate SCDs for the balloon viewing geometry. A direct solar viewing geometry was assumed, even though clouds and multiple scattering processes might change the effective optical paths. This exercise was undertaken for eight flights assuming the presence or absence of a tropospheric BrO contribution of 2 pptv (homogeneously distributed). Data and model simulations from two flights are displayed in Figure 5. Good agreement between measurements and the model was obtained for the 0 pptv case. For the assumed 2 pptv case, significant deviations occur at balloon altitudes below 10 km. Assuming that the AMF are accurate then this indicates that the tropospheric BrO mixing ratios are smaller than 2 pptv, or that they have a variable distribution. It is possible that the tropospheric BrO has a strong diurnal variation with very low mixing ratios at SZA>80°. However this would be in contradiction with observations of high BrO by GOME in February *(i)*, when the satellite overpass takes place at SZA>80° at 68°N. Further radiative transfer calculations have to be carried out to confirm the AMF assumptions and the conclusion.

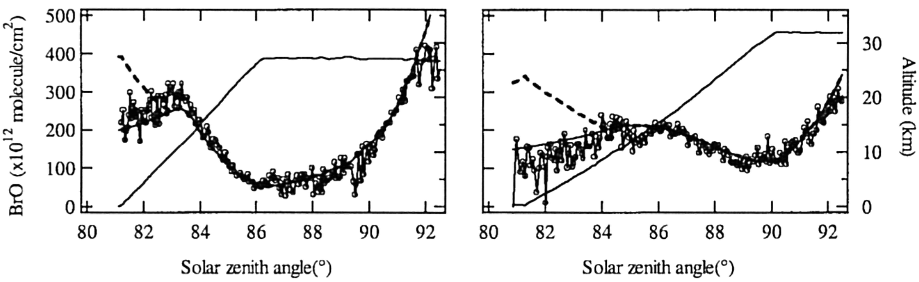

Figure 5. BrO slant columns during balloon flights on 02/03/98 at 69°N (left) and 11/08/98 at 68°N (right) together with balloon altitude. Solid line: model simulation for 0 pptv, dashed line: simulation for 2 pptv tropospheric BrO.

4. Conclusions

The SAOZ/GOME comparison implies tropospheric BrO vertical columns of 1-2*10^{13} molecules/cm^2, corresponding to mixing ratios of 1-2 pptv (if distributed homogeneously). The examination of the influence of clouds on ground-based observations confirms these results. From a study on a cloud-free day a tropospheric mixing ratio of about 3 pptv is estimated. Balloon-borne slant column measurements performed during balloon ascent suggest that tropospheric BrO mixing ratios are smaller than 2 pptv at higher solar zenith angles. Most of the measurements agree if a strong diurnal variation of tropospheric BrO is assumed. Further studies are being undertaken to resolve this issue.

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