SCIAMACHY VALIDATION USING GROUND-BASED DOAS MEASUREMENTS OF THE UNIVERSITY OF BREMEN BREDOM NETWORK

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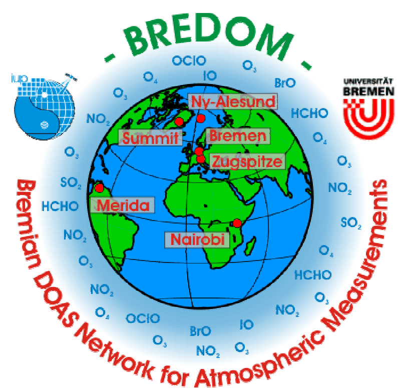
ABSTRACT

A network of ground-based UV/visible DOAS instruments operated by the University of Bremen is used to validate total column measurements from the SCIAMACHY instrument on ENVISAT from the tropics to the Arctic. Measurement quantities include vertical columns of O_3, NO_2, and BrO. Depending on atmospheric conditions, also slant columns of OClO (during chlorine activation) and SO_2, HCHO or IO can be measured. In addition to the standard zenith-sky measurement, all instruments perform off-axis horizon measurements that provide information on the tropospheric burden of the observed species. This quantity will also be used for validation once tropospheric products become available from SCIAMACHY.

In this paper, the instruments and their operation is briefly described as well as the analysis procedures used. Preliminary validation results using SCIAMACHY ozone columns from the Meteo product and NO_2 columns from a DOAS analysis performed on SCIAMACHY lv1 data at the University of Bremen are also presented. Although still little data is available at the time of writing, the first comparisons show the high potential of the SCIAMACHY measurements.

1. THE BREDOM NETWORK

The BREDOM is a network of high quality UV/visible spectrometers for atmospheric observation that is currently set up by the University of Bremen, Germany. The aim is to provide long-term, continuous measurements of a number of stratospheric and tropospheric species at latitudes ranging from the Arctic to the equator. The instruments used for the network are NDSC-qualified [10], and close co-operation exists with the NDSC and other DOAS networks (from the University of Heidelberg, IASB, NIWA) and the SAOZ network of the CNRS, adding to the global atmosphere observation system.



The stations of the BREDOM are given in Table 1. Currently (December 2002), only the instruments in Ny-Alesund, Bremen and Nairobi are operational; the instruments for Summit and Merida are ready and will be shipped to the stations in 2003.

The distribution of the stations is designed to provide a latitudinal cross-section covering Arctic, mid-latitude and tropical regions. This is particularly useful for satellite validation as in the case of SCIAMACHY, as a broad range of atmospheric situations (summer/ winter; high / low ozone, NO_2, H_2O; vortex / non vortex conditions; changing albedo, cloud cover, ...) and also of different measurement conditions (high / low solar elevation) is covered. In addition, the network is also well suited for studies of stratospheric ozone loss, bromine loading and tropospheric pollution. In particular in tropical regions, biomass emissions and lightning produced NO_x as well as stratosphere troposphere exchange leads to large tropospheric ozone concentrations. In order to study this phenomenon more closely, a second tropical station will be added an high altitude site in Venezuela where depending on wind direction clean or polluted air masses in the free troposphere can be probed.

One of the lessons learned from the validation of GOME data is the need for measurements at low latitudes [6, 7, 13]. Compared to Northern mid-latitudes and the polar regions, the density of measurement stations at low latitudes is low and routine zenith-sky measurements of halogen oxides and other minor trace species are not performed. However, such

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measurements are needed for several reasons: First of all, the tropical regions are of paramount importance for the atmosphere as a whole as most of the vertical transport to the stratosphere takes place at these latitudes. Therefore, satellite measurements at low latitudes are of particular interest, and validation of these data has a high priority. In addition, the specific measurement conditions at these latitudes, namely high sun and small light path through the atmosphere are particularly challenging for the satellite instrument. If these data can be validated with good accuracy, many error sources can be minimized that are also important for data taken at other latitudes. For example, GOME measurements of minor trace species are subject to varying offsets that could be corrected if appropriate ground-truth were available at low latitudes. By extending the DOAS network to tropical regions, these missing data will soon be available and will provide an improved validation for SCIAMACHY and also GOME data.

Station	Latitude	Longitude	Status
Ny-Ålesund (i)	79°N	12°E	operational since spring 1995
Summit	72°N	38°W	installation spring 2003
Bremen	53°N	8°E	operational since spring 1993
Zugspitze	47°N	11°W	in planning phase
Merida (ii)	8°N	71°W	installation summer 2003
Nairobi (iii)	1°S	36°E	operational since summer 2002

Table 1: The BREDOM network for SCIAMACHY validation.
(i) in co-operation with AWI Bremerhaven
(ii) in co-operation with Unversidad de los Andes
(iii) in co-operation with UNEP

2. INSTRUMENTS AND DATA ANALYSIS

The instruments used in the BREDOM network are high quality UV/visible DOAS instruments sequentially viewing the zenith sky and the horizon under several elevation angles [1,2,4,9,12]. The main component is a grating spectrometer equipped with a 2-dimensional CCD detector. Light is transmitted from a simple telescope to the spectrometer through a quartz fibre bundle that also converts the spherical aperture of the telescope to the rectangular slit of the spectrometer. In addition, the quartz fibre bundle efficiently de-polarizes the light, an important quality as polarisation of skylight is changing during the day and grating spectrometers have different sensitivities towards parallel and perpendicular polarised light.

The telescope used is shown in Figure 1 and consists of two viewing ports, one to the zenith sky and another towards the horizon. A rotating mirror can be moved into the field of view of the zenith viewing fibre, directing the view towards the horizon at elevation angles between 0° and 30°. Usually, a sequence of 4 different horizon measurements and one zenith sky measurement is taken, each measurement averaging over roughly 1 minute, adding up to a total of 5 minutes for each measurement cycle. However, the instrument is fully programmable and other sequences can be selected for specific situations.

To assure high quality measurements, the instruments have to be kept as stable as possible and be calibrated on a regular basis. To provide calibration, the BREDOM instruments are equipped with a HgCd spectral line lamp and a tungsten lamp in the telescope. Each night, calibration measurements are performed automatically, providing data for monitoring of system performance (spectral alignment, spectral resolution, radiance throughput) and for the correction of instrument parameters in the data analysis (slit function, pixel-to-pixel variation). The spectrometer is housed in an insulated box that is maintained at constant temperature throughout the measurements; the CCD detector is cooled to provide low dark signal. As the telescope is usually situated outside of a building, it is heated to guarantee motor operations and to avoid accumulation of ice and snow on the optical ports.

Operation of the instruments is fully automated and data acquisition is controlled by a computer as a function of time and solar elevation. Data collection as well as instrument control is performed via internet connection. Depending on the location, computer time is adjusted via the internet of from a built in GPS receiver. Except in the case of hardware failure, the instruments run without maintenance for many months.

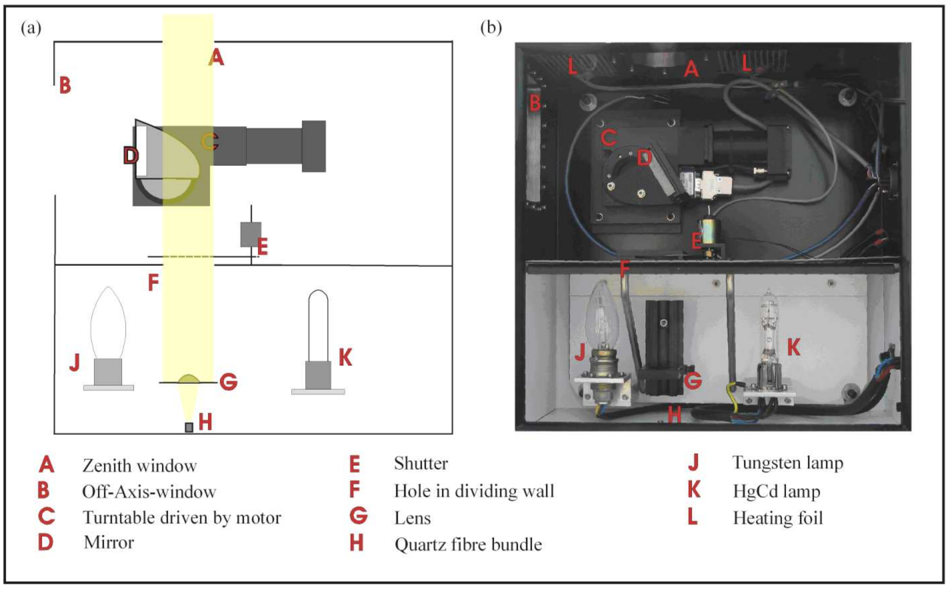

Figure 1: Schematic (left) and photo (right) of the telescope used by the BREDOM instruments

The basic quantity measured by the instruments are spectra between 320 and 600 nm at a spectral resolution of 0.5 to 1.5 nm. Measurements are taken at different solar elevations throughout the day and quasi simultaneously at different viewing angles. From the spectra, optical densities of a number of absorbers in the UV and visible spectral region can be retrieved using the Differential Optical Absorption Spectroscopy (DOAS) technique. Basically, the absorption at different wavelengths is compared and related to the difference in absorption cross-section of atmospheric absorbers. By using many wavelengths, several absorbers can be retrieved at the same time. In order to apply the DOAS retrieval, a background spectrum has to be selected to compensate for the strong Fraunhofer lines that are evident in solar radiation and also in scattered sun light. This background spectrum is either a zenith-sky measurement taken at high sun with the same instrument, or a measurement taken at the same time but in a different viewing direction. The result of the DOAS analysis is a quantity called slant column that corresponds to the integrated amount of the absorber along the (mean) light path through the atmosphere. If the background spectrum also contains atmospheric absorptions, the differential slant column, that is the difference in absorption between the two measurements is retrieved.

To convert this quantity into a geometry independent column, a radiative transfer model is used to simulate the light path through the atmosphere for a given situation, and to compute an enhancement factor (airmass factor) that provides the ratio between the measured slant column and the vertical column that is the absorber concentration integrated vertically from the surface to the top of the atmosphere. In this step, an assumption must be made on the vertical distribution of the absorber in the atmosphere, and depending on wavelength and solar elevation, the results do depend on the a priori information. In the case of ozone analysis in the UV at high latitudes, the dependence is large enough that for a final data evaluation, air mass factors are computed using vertical profiles from ozone sonde measurements close in time and space. At other stations, the variability of the ozone profiles is smaller and also the tropopause height is larger, decreasing the sensitivity of the results on the assumed vertical distribution. Therefore, at these locations, air mass factors calculated from ozone sonde climatologies are used in the analysis.

In the case of NO_2, the profile sensitivity is much lower as the NO_2 maximum is higher in the atmosphere. Therefore, and also because of lack of independent NO_2 profile measurements, climatological NO_2 profiles are used for the calculation of NO_2 airmass factors. In contrast to ozone, NO_2 concentrations in the boundary layer can be very large in polluted regions, and in fact often the amount of NO_2 in the troposphere is larger than that in the stratosphere. To minimise the effect of tropospheric NO_2 on the retrieved column, NO_2 is analysed at low sun only, and in addition, the temperature dependence of the NO_2 absorption cross-section is used to correct for tropospheric absorption. Still, days with large pollution and heavy clouds sometimes have to be excluded from the analysis to guarantee high data quality.

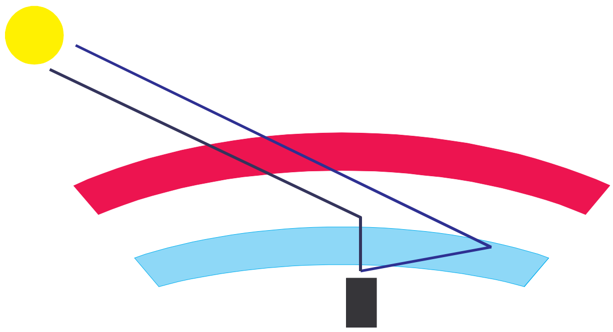

Fig 2: Sketch of the measurement geometry used by the BREDOM instruments. With a scattering height in the middle troposphere, the zenith viewing measurement (black) is weighted towards the stratosphere, whereas the horizon viewing measurement (blue) has a large sensitivity to the layers close to the ground. In first approximation, the stratospheric light paths are the same for both measurement geometries, enabling the boundary layer concentrations to be derived.

One particular new feature of the DOAS instruments used in this study is the capability to observe not only the zenith-sky, but also the horizon. While the standard viewing geometry is heavily weighted towards stratospheric absorbers, the horizon viewing mode is most sensitive to absorptions in the boundary layer (see sketch in Fig. 2) . By combining results from the two measuring geometries and assessing the light path from O_4 absorptions, tropospheric columns can be derived. These columns are well suited for the validation of the tropospheric columns derived from SCIAMACHY measurements by comparing nadir columns with integrated stratospheric profiles once they become available. As a demonstration of the technique, measurements for a clear day in Bremen have been analysed using two different assumptions [2]: a) enhanced NO_2 in the free troposphere and b) enhanced NO_2 in the boundary layer. As can be seen from Fig. 3, measurements taken under different elevation angles give strongly disagreeing values with the first assumption, but nicely fall into line when enhanced boundary layer concentrations are assumed. When this approach is further refined, the vertical distribution of the NO_2 total column in the stratosphere, free troposphere and the boundary layer can be estimated, and two columns, a stratospheric and a tropospheric be determined. Similar results can also be derived for BrO [4] and HCHO [3] in the case of enhanced boundary layer values.

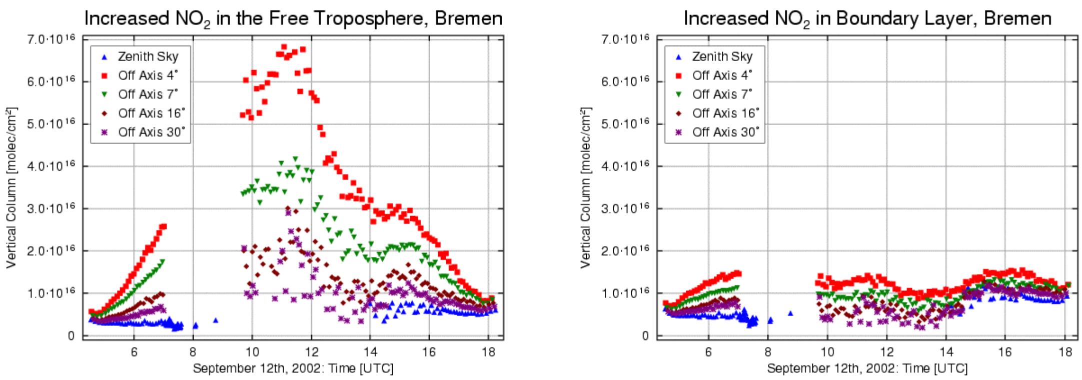

Fig 3: Analysis of BREDOM NO_2 measurements in Bremen on 2002/09/12. The data have been analysed using two different assumptions: enhanced NO_2 in the free troposphere (left) or enhanced NO_2 in the boundary layer (right). As can be seen, only the latter assumption is compatible with the results from all viewing directions. using this approach, a rough estimate on the vertical distribution of the absorbers can be derived.

3. FIRST RESULTS

In a very first attempt to validate SCIAMACHY total ozone products, the SICMACHY meteo-product was used to extract ozone columns above all three operational BREDOM stations and then compared to the ground-based measurements. The reason for using meteo-data instead of standard lv2-products is the availability of data and also the fact, that some problems present in the lv2 products were already fixed in the meteo-files. Results from the intercomparison are shown in Fig. 4.
As can be see from the plots, SCIAMACHY ozone columns are too low by roughly 10% at all stations. The latitudinal variation and also the time evolution (see Bremen data) is well reproduced, and the scatter of the values is a few DU as expected. Recently, one reason for the low values has been identified (an inappropriate slit function was used) and it is expected, that the agreement improves considerably once this problem is resolved. However, the results already show the high potential of the SCIAMACHY ozone measurements.

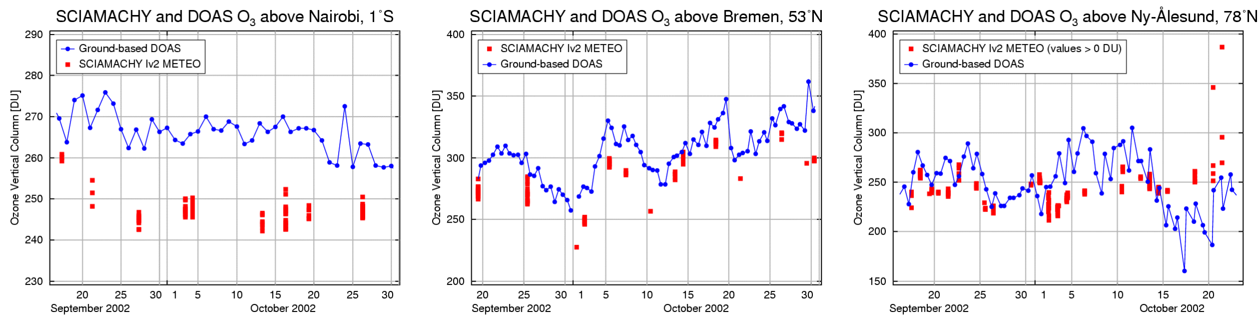

Fig. 4: Comparison of ozone vertical columns measured by the ground-based zenith-sky DOAS instruments and the SCIAMACHY lv2 METEO data product for different latitudes. It can clearly be seen, that the ozone variability is nicely reproduced by SCIAMACHY, but there is an offset with respect to the ground-based measurements. Please note that DOAS measurements in Ny-Alesund have larger errors in October as a result of low light levels.

As no NO_2 columns are included in the SCIAMACHY meteo product, measurements from the BREDOM network were compared to the scientific NO_2 product retrieved from SCIAMACHY lv1 spectra at the University of Bremen [5]. This NO_2 product was derived applying the DOAS analysis developed for the GOME instrument to SCIAMACHY spectra. Starting from lv1 data with only the spectral calibration and dark signal correction applied, NO_2 slant columns are retrieved in a non-linear fit using the wavelength window 425 - 450 nm. In addition to NO_2, the absorptions of O_3, O_4, H_2O and the Ring effect are taken into account, using SCIAMACHY FM reference spectra where possible and a Ring spectrum calculated with the radiative transport model SCIATRAN [11]. Prior to the fit, the wavelength axis of the background spectrum is aligned with a Fraunhofer atlas. In contrast to the standard GOME analysis, no solar spectrum was used as a background, as the SCIAMACHY solar reference measurements provided with the operational data still suffers from unresolved problems. Instead, an arbitrarily selected measurement over the Pacific is used, which leads to an unknown offset in the resulting slant columns that has been accounted for by using the slant column measured by GOME at this location. Once the slant columns are retrieved, they are converted to vertical columns using an airmass factor calculated by SCIATRAN assuming a US standard atmosphere. While this a priori assumption is acceptable in most cases, it will introduce errors at high latitudes in particular in winter.

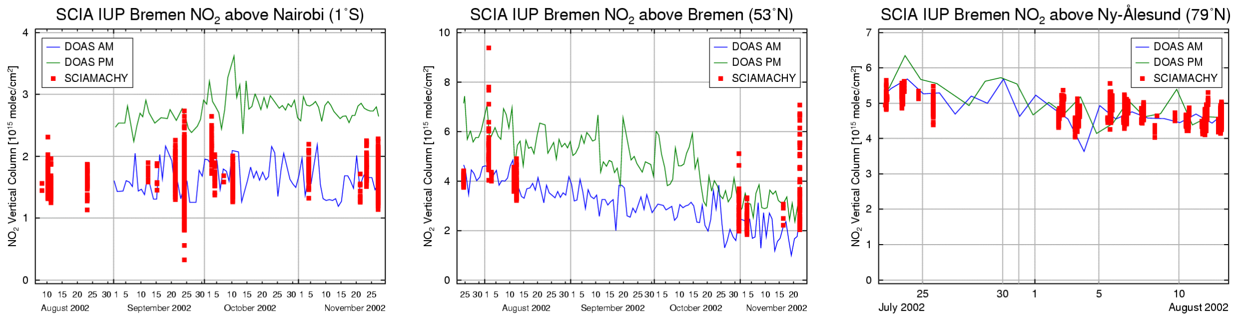

Fig. 5: Comparison of NO_2 vertical columns measured by the ground-based zenith-sky DOAS instruments and the values derived from SCIAMACHY lv1 data for different latitudes. The absolute values and the variability are nicely reproduced. The higher scatter in Bremen is the signature of tropospheric pollution that is corrected for in the ground-based measurements. The number of points is limited by the current availability of SCIAMACHY lv1-data.

The results of the comparison between SCIAMACHY NO_2 and the BREDOM measurements are shown in Fig. 5. As can be seen, both the absolute values and the variation with time is well captured by the SCIAMACHY data at all stations. For the ground-based measurements, morning and evening columns are given as stratospheric NO_2 has a pronounced diurnal variation due to photolysis of N_2O_5. An exception is the situation during polar day where the diurnal variation is relatively small and sampled several times by the satellite instrument as a result of the special viewing geometry at high latitudes. The time of the ENVISAT overpass is close to 11:00 AM in most parts of the world, and a more detailed study of the diurnal variation of stratospheric NO_2 and the viewing geometry of the ground-based instruments shows, that GOME and SCIAMACHY measurements should be close to the AM measurements of zenith-sky viewing instruments. As can be seen from Figure 5, this is in deed the case. An exception are the measurements in Bremen, where SCIAMACHY NO_2 shows a large scatter with some very large values as a result of tropospheric pollution. As pointed out earlier, the zenith-sky measurements have a rather low sensitivity towards tropospheric absorptions, and in addition a correction is applied based on the temperature dependence of the NO_2 cross-section to further reduce the impact of boundary layer NO_2. In contrast, the SCIAMACHY measurements are sensitive to tropospheric NO_2 in the absence of clouds, and in fact can be used to derive global tropospheric NO_2 fields.

4. SUMMARY AND CONCLUSIONS

Measurements from the BREDOM network have been used for a first validation of ozone and NO_2 columns from the SCIAMACHY instrument on ENVISAT. Ozone columns from the SCIAMACHY meteo products are found to be low by roughly 10% in agreement with other studies, probably because of the use of an inappropriate slit function in the operational product. Both the latitudinal variation and the time evolution of ozone columns is well reproduced by the satellite data, and the scatter of the measurements is small, and after correction of the problems still present in the operatinol processing, the quality of the SCIAMACHY ozone product is expected to be similar to that of GOME.

The scientific SCIAMACHY NO_2 product developed at the University of Bremen shows excellent agreement with the ground-based morning values measured at the BREDOM stations with the exception of some large values above Bremen, that are the signature of tropospheric pollution. This demonstrates the high quality of the measurements and the potential to derive a high quality NO_2 product from SCIAMACHY spectra.

In general, it has been demonstrated that the network of ground-based DOAS instruments is well suited for validation of SCIAMACHY column measurements, but current results are still very preliminary as available data from SCIAMACHY are sparse and data distribution and the operational processors not fully functional. SCIAMACHY results from other species (BrO, HCHO, OClO) will be validated as soon as they become available.

5. ACKNOWLEDGEMENTS

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