Atmos. Chem. Phys. Discuss., 6, 9273–9296, 2006
www.atmos-chem-phys-discuss.net/6/9273/2006/

Atmospheric Chemistry and Physics Discussions

Measurements of O_3, NO_2 and BrO at the Kaashidhoo Climate Observatory (KCO) during the INDOEX (INDian Ocean EXperiment) Campaign using ground based DOAS (Differential Optical Absorption Spectroscopy) and satellite based GOME (Global Ozone Monitoring Experiment) data

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Received: 24 July 2006 – Accepted: 15 September 2006 – Published: 26 September 2006

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Abstract

The INDian Ocean EXperiment (INDOEX) was an international, multi-platform field campaign to measure long-range transport of air masses from South and South-East-(SE) Asia towards the Indian Ocean. During the dry monsoon season between January and March 1999, local measurements were carried out from ground based platforms and were compared with satellite based data. The objective of this study was to characterise stratospheric and tropospheric trace gas amounts in the equatorial region, and to investigate the impact of air pollution at this remote site. For the characterisation of the chemical composition of the outflow from the S-SE-Asian region, we performed ground based dual-axis-DOAS (Differential Optical Absorption Spectroscopy) measurements at the KCO (Kaashidhoo Climate Observatory) in the Maldives (5.0° N, 73.5° E). The ground based dual-axis-DOAS measurements were conducted using two different observation modes (off-axis and zenith-sky). This technique allows the separation of the tropospheric and stratospheric columns for different trace gases like O_3 and NO_2. These dual-axis DOAS data were compared with O_3-sonde measurements performed at KCO and satellite based GOME (Global Ozone Measuring Experiment) data during the intensive measuring phase of the INDOEX campaign in February and March 1999. From GOME observations, tropospheric and stratospheric columns for O_3 and NO_2 were retrieved. In addition, the analysis of the O_3-sonde measurements allowed the determination of the tropospheric O_3 amount. The comparison shows that the results of all three measurement systems agree within their error limits. During the INDOEX campaign, background conditions were observed most of the time, but in a single case an increase of tropospheric NO_2 during a short pollution event was observed and the impact on the vertical columns was calculated. In the GOME measurements, evidence was found for large tropospheric contributions to the BrO budget, probably located in the free troposphere and present throughout the year. The latter has been investigated by the comparison of satellite pixels influenced by high and low cloud conditions based on GOME data which allows the determination of the detection limit of tropospheric

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6, 9273–9296, 2006

BrO columns.

1 Introduction

The INDOEX (INDian Ocean EXperiment) project was an international scientific experiment with the main objective to study natural and anthropogenic climate forcing by aerosols and feedbacks on regional and global climate (Ramanathan et al., 2001). INDOEX field studies were carried out over the tropical Indian Ocean, which provides a unique natural laboratory for aerosol studies at the crossing point of pristine air masses from the southern Indian Ocean including Antarctica and rather polluted air masses from the Indian subcontinent. Regional consequences of global warming likely depend on the potentially large cooling effect of aerosols. Therefore, during the intensive field campaign of INDOEX, data were collected from airborne, ship-borne, ground - and satellite based instruments to analyse aerosol composition, atmospheric trace gases, and the long range transport of air pollution from SE Asia towards the Indian Ocean during the dry monsoon (Lelieved et al., 2001). The Kaashidhoo Climate Observatory (KCO), established by the Maldives government and the Scripps Institution of Oceanography is located in the tropics (5.0° N, 73.5° E). The KCO played a central role in the INDOEX campaign (Ramanathan et al., 2001). At KCO measurements with O_3-sondes and in-situ measurements of O_3, carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), CFCs, other chlorinated gases, nitrous oxide (N_2O) and sulfur hexafluoride (SF_6) were carried out alongside meteorological measurements of wind speed and direction, dry air temperature, relative humidity, atmospheric pressure and rain amount during the intensive INDOEX campaign. During INDOEX, the Institute of Environmental Physics and Remote Sensing of the University of Bremen, performed remote sensing measurements using the technique of dual-axis-DOAS (Differential Optical Absorption Spectroscopy) (off-axis and zenith-sky mode) at KCO. This instrument was a precursor of the multi-axis DOAS instrument described in (Wittrock et al., 2004) and (Heckel et al., 2005). Similar set-ups have been developed by other

groups (Hoenninger et al., 2002), (Roozendael et al., 2003). Simultaneous measurements of zenith-sky and off-axis scattered sunlight enables stratospheric and tropospheric amounts of atmospheric gases to be distinguished. Specifically the total and tropospheric column amounts of ozone (O_3), nitrogen dioxide (NO_2), as well as tropospheric amounts of BrO above KCO were to be investigated. The results of our O_3 and NO_2 measurements were to be compared with those from GOME (Global Ozone Monitoring Experiment) (Burrows et al., 1999). The subject of this contribution is the validation and synergistic use of ground based and remotely sensed GOME data, as well as O_3-sonde measurements above the Indian Ocean during INDOEX.

2 Experimental set-up

2.1 Ground based DOAS measurements

During the intensive INDOEX campaign, DOAS measurements were carried out at KCO using two selected viewing geometries (dual technique: zenith and off-axis) (see Fig. 1). The dual-axis-DOAS-instrument is comprises of a Czerny Turner spectrometer (ARC 500, Acton Research Coporation) and a custom made telescope. High temporal resolution was achieved by switching between zenith-sky and 2° above the horizon direction within a short time interval (typically a few minutes). We can therefore assume essentially no change in the vertical column amount between the two recordings. For the observations at the elevation angle of 2°(angle between the horizontal direction and the viewing direction of the telescope) most of the light is scattered into the telescope from the air close to the surface and a height of 2 km resulting in a higher sensitivity to the lower atmosphere compared to the observation of zenith scattered sunlight (90°), which has a comparatively short path through the troposphere. These measurements were carried out each day between sunrise (average over the morning measurements = a.m.-value) and sunset (average over the evening measurements = p.m.-value) in the wavelength region of 327–492 nm with a spectral resolution of 0.5 nm. For this study,

spectra in the wavelength region of 435–481 nm were analysed applying the DOAS technique (Platt, 1994), (Wittrock et al., 2004), (Heckel et al., 2005) to obtain the differential slant column (DSC), the difference between the column amount of the absorber integrated along the light path through the atmosphere and the absorber amount in the background spectrum (SC_0) at the smallest solar zenith angle of O_3 and NO_2. The fifth order polynomial was fitted to the spectrum and absorptions by O_3 (Burrows et al., 1999), NO_2 (Burrows et al., 1998), the oxygen dimer O_4 (Greenblatt et al., 1990) as well as H_2O (Rothman et al., 1992) were taken into account. The effect of Raman scattering (Grainger and Ring, 1962) was compensated by fitting a synthetic Ring spectrum as an additional absorber (Vountas et al., 1998). Total vertical columns of the trace gases O_3 and NO_2 for the location Kaashidhoo can be derived by the division of the slant columns (the SC is the sum of the DSC and the SC_0 in the background spectrum) of the zenith measurements by the air mass factor (AMF) which describes the light path through the atmosphere and is calculated with GOMETRAN/SCIATRAN (Rozanov et al., 1997). Error analysis revealed a maximum relative error of 5% for the total amount for either trace gas retrieval, taking into account the radiative transfer in the atmosphere for the AMF calculation including the assumed shape of the vertical profile of the absorber, the surface albedo, and the aerosol loading.

The total vertical column of the trace gases consists of a tropospheric as well as a stratospheric part. Using both observation modes and appropriate AMF calculations, tropospheric vertical columns and/or profiles can be separated from the measurements (Heckel et al., 2005), (Wittrock et al., 2004), as carried out in this study for the trace gas O_3. The analysis of the tropospheric amount of NO_2 is based on the results of dual-axis-DOAS measurements combined with model assumptions. The following simplifying assumptions were made for the calculation of tropospheric NO_2: (a) a polluted and well mixed air mass (air masses with higher amounts of NO_2 than in the surrounding air) follows the wind from north to south, (b) this air parcel passes both observing modes without any exchange with the environment and (c) the air mass has a rectangular shape with homogenously high amounts of NO_2 (see Fig. 2). With these

assumptions the analysed air parcel is passing first the off-axis observing mode and then the zenith observing direction (see Fig. 2).

2.2 GOME and O_3-sonde measurements

GOME was launched in April 1995 onboard the second European satellite ERS-2 in a sun-synchronous near polar orbit at a mean altitude of 795 km for measuring the sunlight scattered from the Earth's atmosphere and/or reflected by the surface in nadir mode in a wavelength region of 240 to 790 nm with a spectral resolution of 0.2–0.4 nm. The local equator crossing time is 10:30 a.m. With 14 orbits per day, global coverage at the equator is reached after three days for a 960 km swath width (the size of one GOME ground pixel is $40 \times 320\,km^2$) (Burrows et al., 1999). For this study GOME data reprocessed with WFDOAS (Weighting Function Differential Optical Absorption Spectroscopy) Version 1 (Coldewey et al., 2005) and (Weber et al., 2005) were extracted with a maximum distance of 300 km away from the location of interest to derive vertical columns of O_3. The analysis to derive slant columns of NO_2 (Richter and Burrows, 2002) in the 425–450 nm and of BrO in the 344–359 nm wavelength region (Richter et al., 2002a) was carried out using the IUP Bremen DOAS algorithm. The retrieval method is similar to that used for ground-based zenith-sky measurements. For the comparisons with ground based DOAS measurements, only GOME level 1-spectra with a maximum distance of 300 km around Kaashidhoo were extracted. GOME observes both – the troposphere and the stratosphere (Burrows et al., 1999) – and in cloudy conditions it is impossible for GOME to detect the exact influence of trace gases below the cloud layer. For the comparison of the tropospheric column amounts from GOME with ground based DOAS measurements, only GOME-pixels under cloud free conditions (using measurements with a cloud fraction of less than 0.2 as determined by the FRESCO algorithm, Koelemeijer et al., 2001) were included in the analysis. The tropospheric trace gas columns were derived by applying the tropospheric excess method (TEM) or reference sector approach. The TEM is based on the assumption that the columns of stratospheric trace gases such as NO_2

and O_3 are approximately constant at a given longitude. This is a large over simplification but appears to work well for tropical and sub tropical conditions for O_3 and NO_2 and to higher latitudes mainly for NO_2. A number of studies have been published focusing on the retrieval of tropospheric NO_2 (Leue et al., 2001), (Richter and Burrows, 2002), (Martin et al., 2002) and O_3 (Ladstätter-Weißenmayer et al., 2004) from GOME data. The error on the resultant tropospheric O_3 is estimated to be 4 DU (Dobson Units) (Ladstätter-Weißenmayer et al., 2004) and on the tropospheric columns of NO_2 approximately 1.5×10^{15} molecules cm^{-2} (Richter and Burrows, 2002) based on GOME data. During the INDOEX campaign in January to March 1999, in addition to the Vaisala radio sondes (Komhyr et al., 1995) a collection of 55 electrochemical cell (ECC) ozone soundings (Lobert et al., 2002) and (Thompson et al., 2003) were launched about once per day at approximately 14:00 local time (09:00 Universal Time (UT)) at the KCO by the Scripps Institution of Oceanography. In order to obtain the tropospheric vertical columns of O_3, the sonde data were summed up to the height of the tropopause defined in this case study to be at 4 PV (potential vorticity) Units.

3 Results and discussion

3.1 Total column amounts of O_3 and NO_2

The ground based DOAS measurements were used to determine a.m.-, and p.m.-, values for O_3 and NO_2 total columns. The latter were determined from the observations at SZAs between 83 and 90° in the morning and afternoon. The analysis of the zenith-sky ground based measurements of O_3 showed almost no difference between a.m. and p.m. values (Fig. 3a). That means no diurnal variation for the vertical columns of O_3 can be seen during the whole INDOEX campaign. A mean value of 263 DU was determined using both a.m. and p.m. data. Deviations of up to 10 DU from the mean value which can be attributed to the transport of different air masses, are observed. Comparison of these data with the results of satellite based GOME measurements

shows agreement within 3% for the total columns. This value is smaller than the combined errors of the ground based measurements and the satellite based measurements of 5%. Figure 3b shows the a.m. and p.m. values for NO_2 vertical columns (given in molecules cm^{-2}) from the ground based DOAS measurements compared to the GOME results. The difference between the a.m. (around 06:30 a.m.) and the p.m. (around 17:30 p.m.) values of the ground based measurements was significant for NO_2. The p.m. values were almost a factor of 1.7 higher than the a.m. values. This behaviour is attributed to the photolysis of N_2O_5 in the stratosphere. The influence of the diurnal cycle is more intense in the tropics compared to mid-latitudes where the factor is typically 1.4 (Solomon et al., 1987) for ground based measurements. A result of a NO_2 daytime increase of 1×10^{14} cm^{-2}/h can be calculated from our ground based measurements. A similar result is obtained comparing ground based measurements carried out in the morning and GOME data. The retrieved a.m. columns of NO_2 show roughly 25% lower values compared to the GOME results. In comparison to the ground based measurements, the total columns of NO_2 from GOME are 4×10^{14} cm^{-2} (mean value) higher because of its later overpass time of 10:30 a.m. at KCO compared to the ground based measurements carried out 4 h earlier. While an increase of 1×10^{14} cm^{-2}/h can be calculated comparing ground based a.m. and p.m. results as well as ground based a.m. and GOME data, the output of a photochemical model (Sinnhuber et al., 2005) based on the chemistry scheme from the SLIMCAT model (Chipperfield, 1999), with reaction rate constants taken from the JPL-2002 recommendations (Sander et al., 2002) (see Fig. 3c) shows an increase of only 3×10^{13} cm^{-2}/h caused by the already mentioned diurnal variations in stratospheric NO_2.

3.2 Tropospheric column amounts of O_3, NO_2 and BrO

The second scientific aim of this study was to determine the influence of pollution events on the tropospheric column amounts of O_3 and NO_2.

Figure 4 shows the comparison between the tropospheric column of O_3 determined

as described above from GOME and that derived from the ozone sonde measurements. The results of tropospheric amounts of O_3 from both instruments are in the range of background conditions because the tropical Indian Ocean is dominantly influenced by pristine air masses from the southern Indian Ocean including Antarctica during this time of the year. The results of two selected case studies, depicted in red in Fig. 4, show lower tropospheric ozone columns for the ground based DOAS data compared to the results of either GOME (40%) or ozone sonde measurements (33%) on these two days, whereas the differences between the sonde data and the remotely sensed data are within a range of 4% (1.2 DU). Taking into account the error bars of both the O_3-sondes and satellite based data, during this episode the two instruments agreed within their error limits (see Fig. 4).

The reason for the differences in the values between ground based, satellite, and ozone sonde measurements is likely a combination of limitations arising from the DOAS ground based measurements. One of the main error sources is poorly known type and amount of aerosol. Following from the high amount of aerosol over the Indian Ocean, the use of the dual-axis-DOAS instrument, in contrast to the MAX (multi axis)-DOAS instrument (Wittrock et al., 2004), as used presently, can introduce large errors. That means, the calculation of the tropospheric AMFs taking into account only two different observing modes (zenith-sky and off-axis) consequently leads to an overall uncertainty of the analysis with estimated errors on the order of 10 DU, a value usually expected for this type of study. Thus, within the combined error limits, the ground based measurements agree with the GOME results. In this study, in addition to the analysis of tropospheric O_3, the influence of tropospheric pollution with respect to NO_2 was determined. Using the high temporal resolution of ground based data, a localised tropospheric NO_2 plume could be observed on 15 March 1999. An increase of the slant column of NO_2 due to polluted air masses was observed in the off-axis mode first. The same polluted air mass was later transported through the zenith-sky telescope's field of view, and the slant columns of the zenith-sky measurements were increasing as well. After the air mass has passed the telescope, the slant columns of both observing modes were

then decreasing back to the background value. Based on the measurements of both observing modes, the increase and the following decrease of the NO_2 slant columns can be determined as a function of time (see Fig. 5a). Using the wind speed in off axis direction (v_W=5 m s^{-1}), as measured at KCO, the elevation angle of the off axis viewing mode (α=2°), the assumed vertical height of the plume (h=s*tan(α)=262 m) with s=7500 m (the horizontal distance of the plume from the measuring point), its horizontal extension in the viewing direction (l=(time of decrease of SC of both viewing modes - time of the first increase of SC of zenith data)*v=900 m) and its horizontal distance from the measuring point at a defined time (see Fig. 2) the tropospheric NO_2 amount during a short pollution event can be obtained.

Dual-axis-DOAS reveals for the 15 March 1999 a maximum concentration of 1–2×10^{11} molecules cm^{-3} (5 ppb) of NO_2 based on the model assumption for a well mixed air mass over the Maldives (see Fig. 5b). On this day the observed total vertical column amount from ground based measurements was 1.8×10^{15} molecules cm^{-2} (a.m. value, (see Fig. 3b). This means using the maximum concentration of 1–2×10^{11} molecules cm^{-3}, a tropospheric vertical column amount for NO_2 of 7.1×10^{15} molecules cm^{-2} was determined. Therefore, on this day an increase by a factor of 4 occurred over Kaashidhoo during this brief pollution event (see Fig. 5b) for the total column amount of NO_2.

Comparing this result with the tropospheric amount of NO_2 measured with GOME for the same day, a difference of 6.1×10^{15} molecules cm^{-2} can be observed (see Fig. 5b). This case study shows that a local increase of tropospheric NO_2 up to 7×1×10^{15} molecules cm^{-2} could be observed with ground based DOAS measurements in the afternoon whereas GOME is measuring 1×10^{15} molecules cm^{-2} at 10:30 over this region, highlighting the importance of different spatial and temporal resolutions of both instruments. Locally observed high values for tropospheric NO_2 can be caused by different sources like combustion processes of anthropogenic or natural origin, such as biomass burning, and lightning discharge (Wayne, 1991). In this particular case, the increase of NO_2 was probably caused by nitrogen oxide (NO_x

= NO + NO_2) emissions from ships (Lawrence et al., 1999; Kasibhatla et al., 2000; Richter et al., 2004; Wittrock et al., 2004). From GOME measurements the mean background tropospheric amount of NO_2 was determined to be 4×10^{14} molecules cm^{-2}, while the mean total column amount of NO_2 during the INDOEX campaign was 1.7×10^{15} molecules cm^{-2}. That means an average tropospheric fraction of 24% under clean air background conditions was observed.

Very large BrO columns have been detected in polar spring in both hemispheres. These events have already been studied and compared with model output, balloon-borne observations and ground based measurements in previous publications (Wagner and Platt, 1998; Richter et al., 1998; Richter et al., 1999; Wagner et al., 2001; Roozendael et al., 1999; Roozendael et al., 2002). They are attributed to boundary layer effects, in which photochemically produced active bromine can temporarily accumulate to high abundances in a shallow surface layer, consuming ozone while not being sequestered in chemically more stable bromine species. The source of BrO has not been identified beyond doubt but active bromine species are presumed to be released either from sea salt, especially in the form of frost-flowers, or from biogenic organo-bromine species. In addition to the large values observed in polar spring, enhanced BrO is also observed globally in GOME data (Hegels et al., 1998; Roozendael et al., 1999; Richter et al., 2002a). This has tentatively been attributed to a significant background concentration of BrO in the free troposphere of several ppt, and direct evidence for BrO in the free troposphere was found by (Fitzenberger et al., 2000). Here, this assumption has been tested by analysing ground-based DOAS measurements and satellite-based GOME data during the INDOEX campaign in the wavelength region 344-359 nm where BrO shows strong absorption features. Since several power failures have had an impact on the stability of the ground based instrument, only a detection limit of 3×10^{14} molecules cm^{-2} was achieved for BrO from our ground based dual-axis-DOAS measurements. Using the difference of low (at 900–1013 mbar) and high cloud (at 0–600 mbar) situations during the INDOEX campaign an upper limit for the free tropospheric column of BrO of 3.8×10^{13} molecules cm^{-2} from GOME pixels

(see Fig. 6) can be observed. Assuming a 4 km thick atmospheric layer at 1000 hPa, 4 ppt of BrO can be calculated for this time period. That means a small amount of BrO is present in the troposphere (Richter et al., 2002a). The value calculated in this study for the amount of tropospheric BrO during the campaign is representative for background conditions as estimated in other studies (Richter et al., 1998; Richter et al., 1999; Wagner and Platt, 1998; Wagner et al., 2001; Pundt et al., 2002; Fitzenberger et al., 2000; Dickerson et al., 1999).

4 Conclusions

During the INDOEX campaign in February and March 1999 on Kaashidhoo, ground based dual-axis DOAS measurements (zenith and off-axis-measurements) were performed in the equatorial region. This method provides information of total as well as on tropospheric column amounts of O_3 and NO_2 and can distinguish between background and polluted atmospheric situations. In addition, an upper limit for the free tropospheric amount of BrO could be given using cloud-affected GOME measurements. The comparison between ground based dual-axis DOAS, GOME (both detecting O_3, NO_2, BrO), and O_3-sonde data demonstrates that all measurement systems agreed within their error limits. The relatively large errors in the ground based data result from both instrumental and theoretical errors related to the assumed homogeneity of the tropical air masses encountered. Differences between ground based and satellite measurements can result from the temporal variability of the atmosphere when measurements are not exactly coincident in time, and from the limited spatial resolution of GOME compared to ground based measurements. This dual-axis-DOAS-technique and in addition the calculation of the different AMFs allowed to calculate around 19 DU for tropospheric O_3 (two case studies on 19 February and 14 March 1999) and 263 DU for the total amount of O_3. The results were compared to satellite based GOME-data and O_3-sondes launched over Kaashidhoo. Differences of up to 40% for the GOME (24.6–32.5 DU) data and of 33% for the O_3-sonde measurements (20.6–36.8 DU) in

the tropospheric part for both case studies and a discrepancy of 3% for the vertical columns for the time period of the INDOEX campaign were obtained. Pollution events such as on 15 March 1999 could be detected by applying both, the zenith-sky and the off-axis mode of the ground based measurements. Therefore, this measurement method permits to differentiate between local tropospheric emissions of, e.g., NO and stratospheric NO_2 columns. A simple model based on a homogenous air mass with enhanced NO_2-loading passing the two different observing modes yields an increase of up to 5–10 ppb in this case study. GOME satellite data from the same day show no significant increase in NO_2, highlighting the strengths and weaknesses of the two different observing systems. While GOME has excellent spatial coverage but limited spatial and temporal resolution and therefore limited sensitivity for the detection of localized events, the ground based measurements with the dual-axis-technique can observe local pollution events on a short time scale at one location. Furthermore, we calculated an upper limit of 3.8×10^{13} molecules cm^{-2} from BrO satellite based measurements for the tropical free troposphere, comparing pixels of GOME measurements under high and low cloud situations, and made possible by the high spectral resolution of GOME. This abundance confirms previous studies (Richter et al., 1998; Richter et al., 1999; Wagner and Platt, 1998; Wagner et al., 2001; Pundt et al., 2002; Fitzenberger et al., 2000; Dickerson et al., 1999).

Acknowledgements. Parts of this work have been funded by the University of Bremen, Germany, the DLR/DARA, the European Community, and the European Space Agency (ESA). We would like to thank H. Altmeyer who carried out with M. Bruns the measurements at KCO. We would like to thank R. B. A. Koelemeijer for providing the FRESCO data. We would like to thank the International sharing steering Group of the INDOEX campaign for great assistance, without which this work would not have been possible. We thank especially the organizers of the INDOEX campaign P. Crutzen and V. Ramanathan, J. Lobert and the whole KCO team during the IFP 1999. We thank in particular R. Dickerson for great help during the campaign. We also appreciate the warm hospitality of the Maldivians on the island Kaashidhoo, Republic of Maldives. Discussions with R. v. Kuhlmann, M. G. Lawrence and A. M. Thompson were very helpful.

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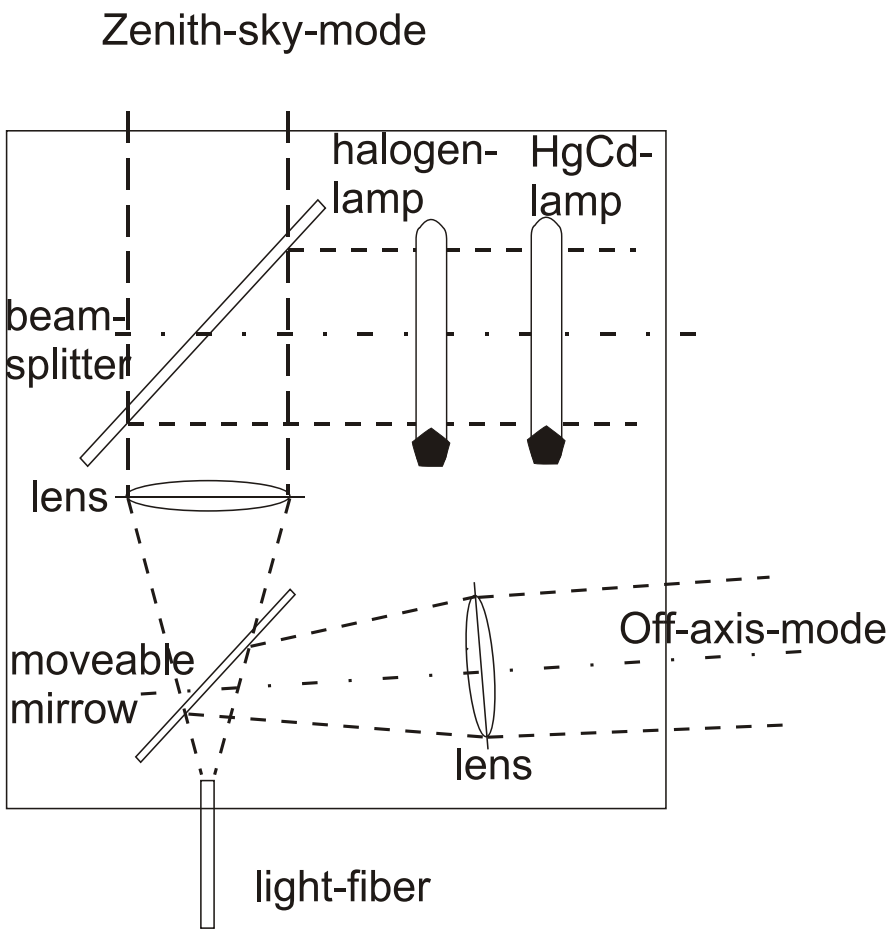

Fig. 1. Experimental setup of the DOAS telescope used in Kaashidhoo from 14 February to 16 March 1999 during the INDOEX campaign.

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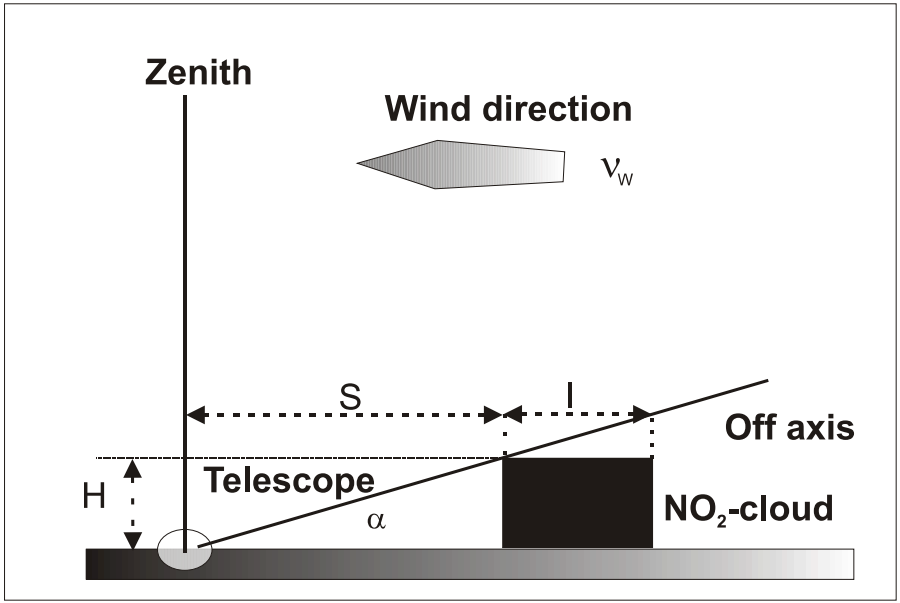

Fig. 2. The conceptual model used for describing a plume with high NO_2 amounts as observed with zenith and off axis geometry.

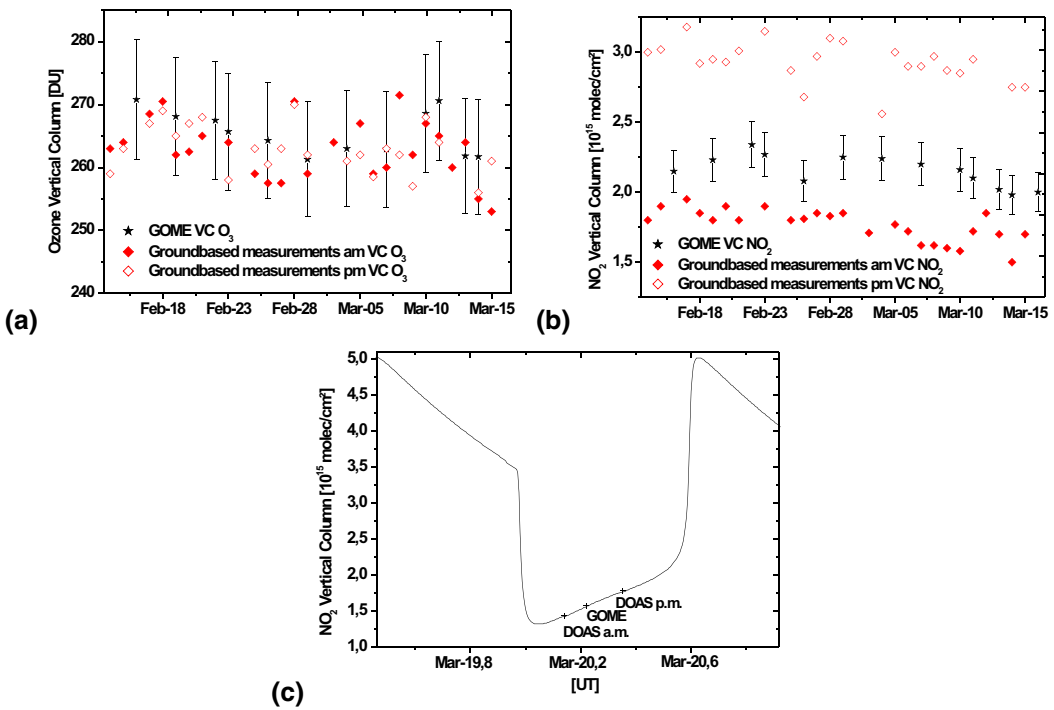

Fig. 3. Total columns of O_3 **(a)** and NO_2 **(b)** measured by the ground based DOAS system and with GOME during the INDOEX campaign from 14 February to 16 March 1999. **(c)** Model calculation of the diurnal cycle of NO_2 for 19 to 20 March 1999 including the measuring time of ground based a.m. and p.m. as well as the overpass time of GOME.

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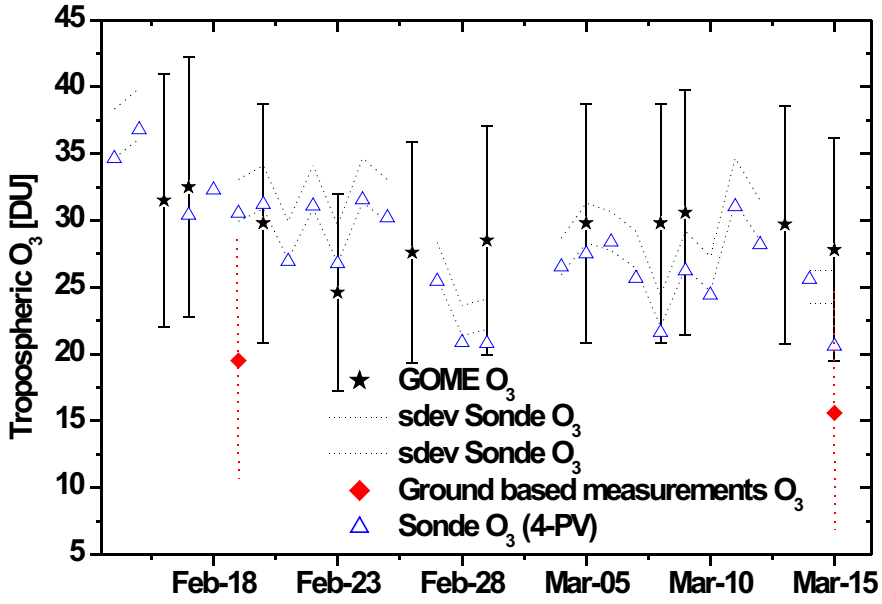

Fig. 4. Tropospheric columns of O_3 measured by the ground based DOAS system compared with the O_3-sondes and with GOME data for the time period of 14 February to 16 March 1999 during the INDOEX campaign.

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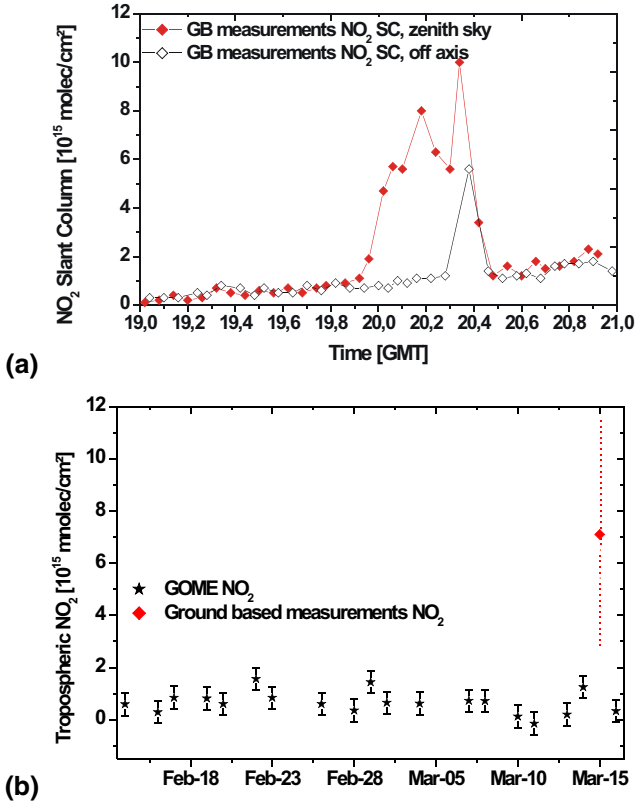

Fig. 5. Tropospheric slant columns of NO_2 **(a)** measured by ground based (GB) DOAS system (off-axis and zenith-sky-data) for March 15, 1999. Tropospheric columns of NO_2 **(b)** measured by ground based DOAS system and compared with tropospheric excess columns of GOME (GOME is only measuring at 10:30 a.m. and therefore not able to observe an enhancement of localised NO_2 in the afternoon) for the time period of 14 February to 16 March 1999 during the INDOEX campaign.

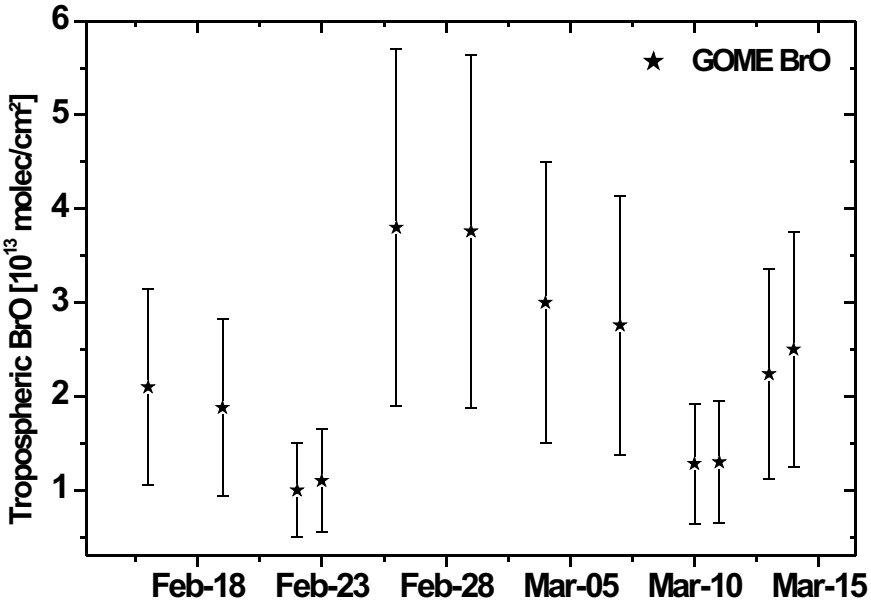

Fig. 6. Free tropospheric columns of BrO calculated from GOME data for the time period of February to March 1999 during the INDOEX campaign using the difference between "low cloud" and "high cloud" observations.