OF TROPOSPHERIC TRACE GASES

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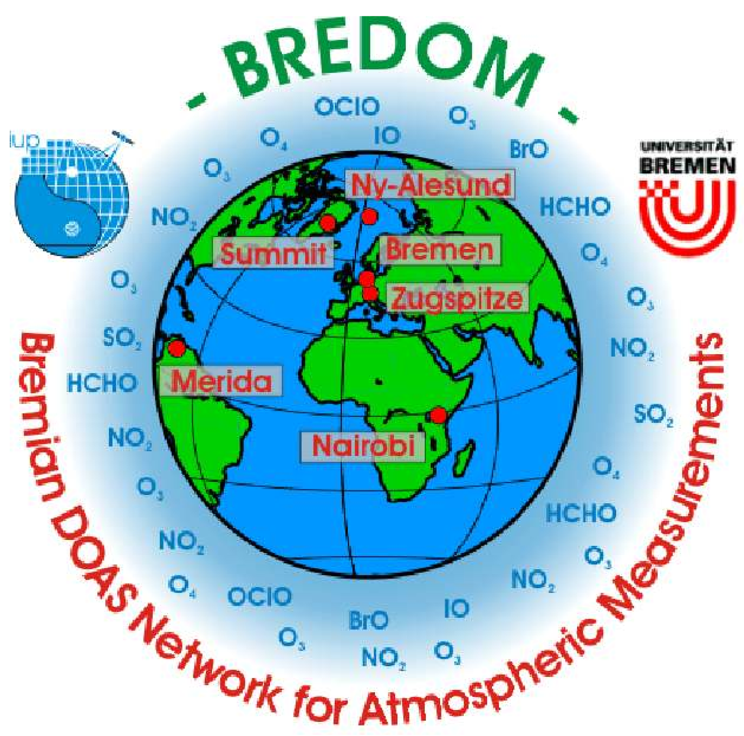
Introduction

The Bremian DOAS network for atmospheric measurements (BREDOM) is a network of high quality UV/visible spectrometers for atmospheric observation that has been 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. This is particularly useful for satellite validation, as a broad range of atmospheric situations (summer/ winter, high/ low ozone, 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 tropospheric pollution (e.g. biomass emissions), because all instruments are equipped with the MAX(multi-axis)-DOAS technique.

An important step in passive remote sensing was the development from ground-based zenith sky observations to multi-axis measurements [1], which has enabled us to validate findings from satellite observations and study the behaviour of important trace gases in the troposphere on a local scale. Applying the Optimal Estimation Method [3] to measured slant columns yield profile information on numerous trace gases in the troposphere [2].



Furthermore the aerosol optical depth and other aerosol features can be retrieved using measurements of the oxygen dimer O_4.

This study shows time series for tropospheric amounts of NO_2, and HCHO for selected BREDOM stations and for different field campaigns. In addition selected data sets have been examined for aerosol properties. These data sets have been compared to other ground-based data.

DOAS Setup

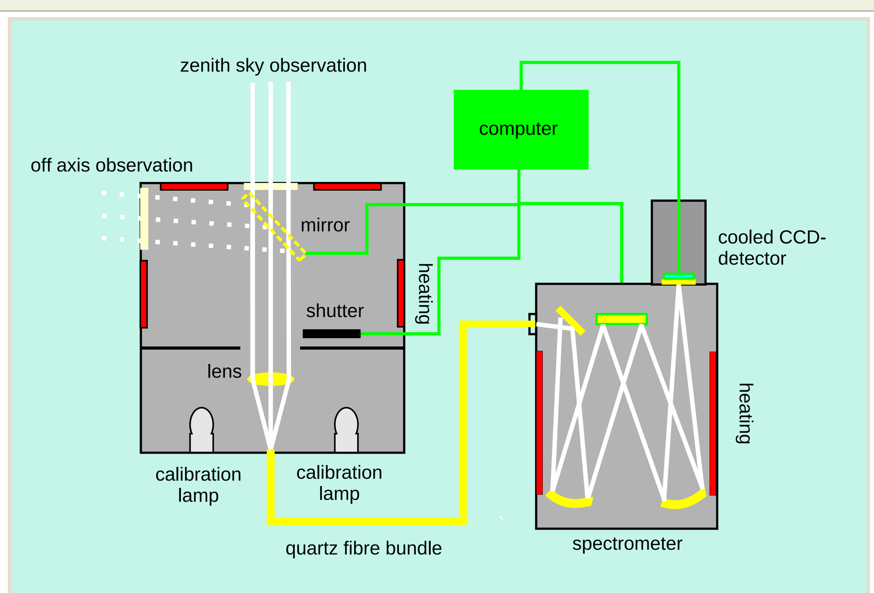


Figure 1 DOAS setup

Sunlight scattered from the sky is collected by a telescope (Fig 1) and transmitted to a Czerny-Turner spectrograph via a depolarizing quartz-fibre bundle. A charge coupled device (CCD) is used as a detector. The pointing of the telescope alternates between zenith and four off-axis directions (0° to 30° above horizon), which provides profile information of the absorbers. The time resolution is usually 5 minutes for a complete measurement cycle. The observation in different lines of sight is realized by a mirror fixed on a revolving table driven by a computer controlled motor. The whole system works automatically and the measurement parameters can be set from Bremen via an internet connection.

Profile Retrieval: BREAM

BREAM: Bremian advanced MAX-DOAS retrieval algorithm

Recent studies [4], [1] have shown that the measured slant column of the oxygen dimer O_4 can be used to derive aerosol information, i.e. the extinction profile and to some extent also the aerosol type in the atmosphere. The knowledge is essential to retrieve the correct amount of the chosen absorber (e.g. HCHO).

In a first step the algorithm uses the radiation transport model SCIATRAN [5] to calculate O_4 slant columns which are compared to the measured ones in order to reduce uncertainties due to aerosols. The extinction profile in its total quantity as well as in its structure (i.e. the height of the boundary layer) is scaled in an iterative process and therewith the slant columns of O_4 are calculated. The quality of the agreement is evaluated by applying two parameters: The correlation between the measurement and the modelled columns (which is mainly influenced by the height of the boundary layer) and the mean deviation of those (mainly modulated by the extinction).

The second step comprises calculation of so called block air mass factors with respect to the chosen absorber using the prior obtained aerosol information. Again SCIATRAN in its full-spherical mode is operated. Block air mass factors are air mass factors which depend on the layer height of the absorber. The overall air mass factor is simply the average of the block air mass factors weighted by the distribution of the trace gas. This concept allows us to describe the relation between the measurement (i.e. the set of slant columns under different elevation angles of the absorber) and the absorber's profile in the atmosphere as a linear system. To solve such a linear system the well-known and in atmospheric chemistry long-established method of Optimal Estimation by Rodgers [3] is applied.

Vertical Resolution

The averaging kernel matrix is a measure for the quality of the retrieval. The trace of the matrix determines the number of pieces of independent information with respect to the height layers, i.e. the degrees of freedom of this measurement. Typical integration times (20 minutes to one hour) together with a moderate aerosol content yields a number of about two to three. The whole range includes results from about 1.5 for low visibility up to about five considering also large solar zenith angles (SZA > 75°). An example for the averaging kernal matrix and the corresponding retrieved profile is shown in Figure 2.

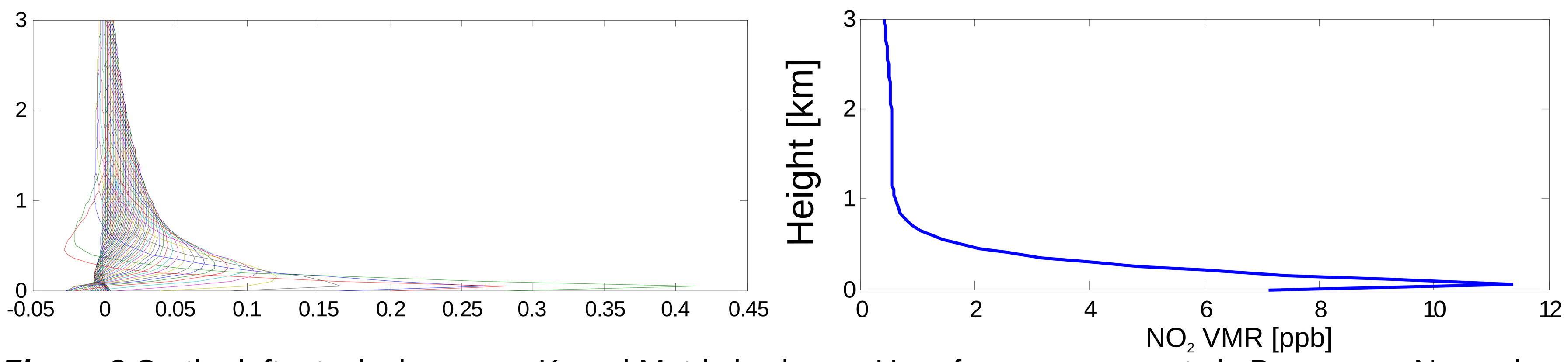


Figure 2 On the left a typical average Kernal Matrix is shown. Here for measurements in Bremen on November 12, 2004. The plot on the right side shows the corresponding NO_2 profile between 7:00 and 7:30.

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Universität Bremen

Alzate (46°N, 9°E)

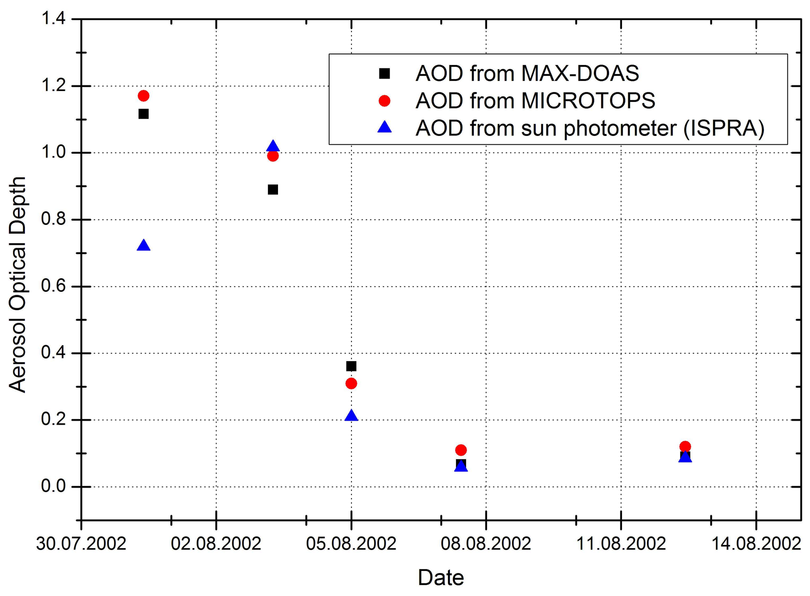


Figure 3 Comparison of different aerosol measurements at Alzate, Italy. The MAX-DOAS results have been retrieved using BREAM.

In summer 2002 and autumn 2003 two campaigns were hold in the Milan aera in the frame of the FORMAT-project. A variety of instruments were installed in three different locations which allows comparison of data obtained by different platforms:

Figure 3 illustrates the quality of the retrieved aerosol data using BREAM. A comparison for the aerosol optical density for three different instruments during the first FORMAT-campaign is shown: Measurements with a sun photometer from the JRC in Ispra as well as with a MICROTOPS-instrument from the IMK-IFU together with the aerosol optical density retrieved from the O_4 measurements with a MAX-DOAS instrument. Excellent agreement is found.

The volume mixing ratio for HCHO for both campaigns is shown in Figure 4. MAX-DOAS results are compared to Longpath-DOAS from the IUP at the University of Heidelberg and Hantzsch from the IMK-IFU.

In general the MAX-DOAS detects higher HCHO VMRs than the two more local instruments. This might be caused by a

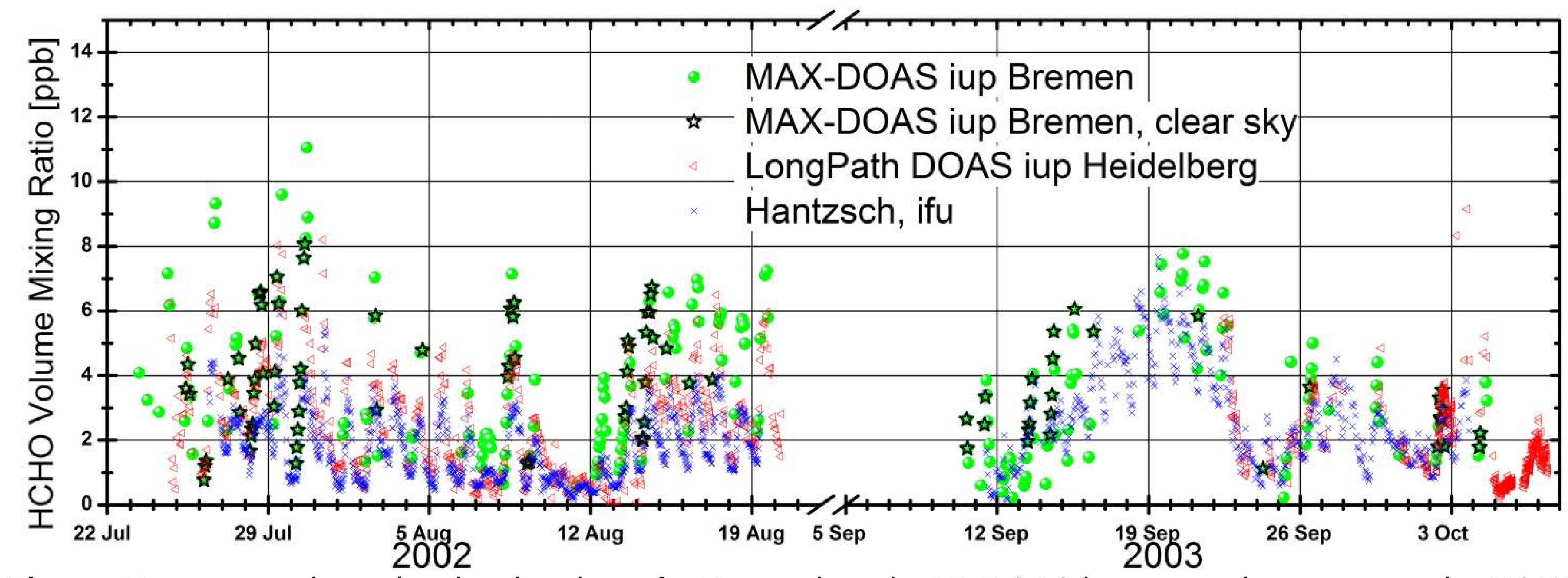
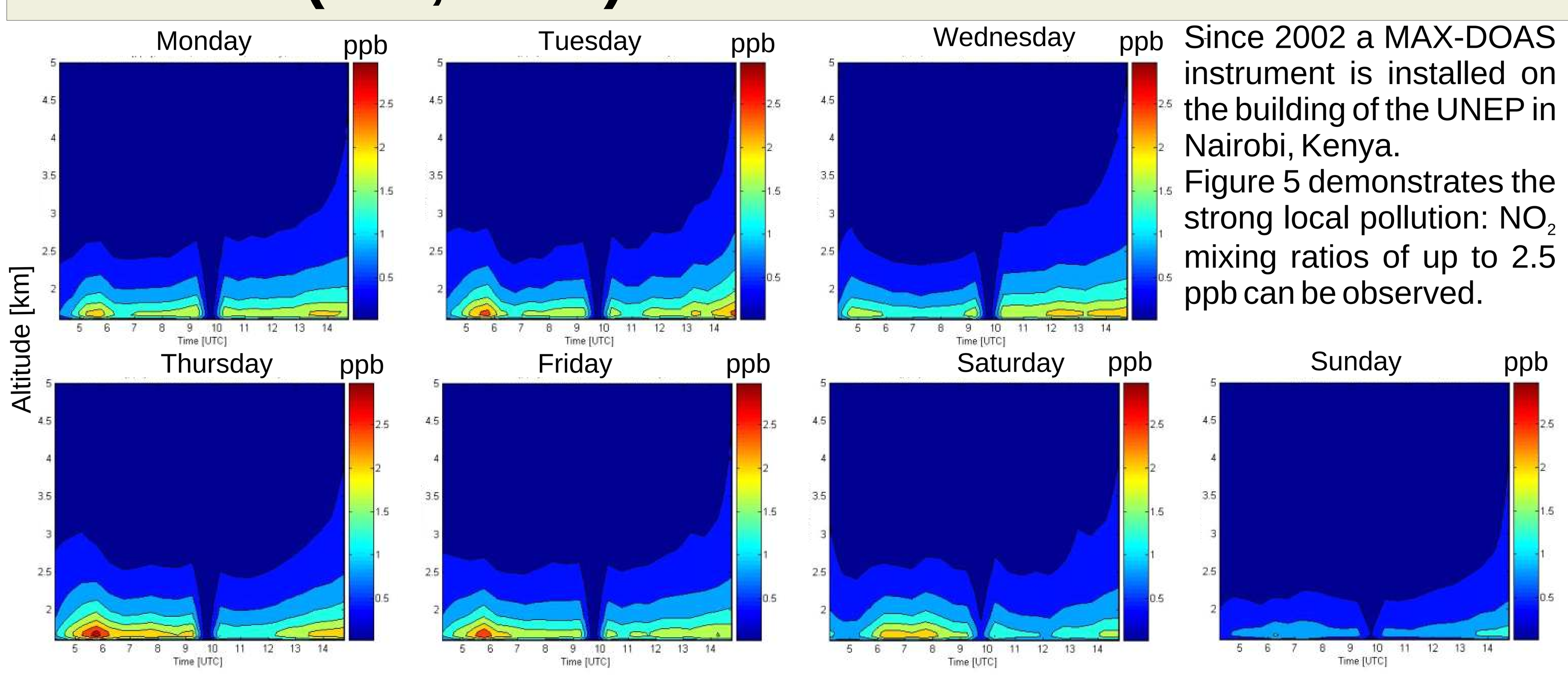


Figure 4 Intercomparison plot showing data of a Hantzsch and a LP-DOAS instrument in contrast to the HCHO mixing ratio derived using BREAM for an atmospheric bottom layer of 150 m height from the MAX-DOAS analysis.

Nairobi (1°S, 36°E)



Since 2002 a MAX-DOAS instrument is installed on the building of the UNEP in Nairobi, Kenya.

Figure 5 demonstrates the strong local pollution: NO_2 mixing ratios of up to 2.5 ppb can be observed.

Figure 5 NO_2 profiles for one weak retrieved using BREAM. A clear weekend-effect can be observed: On Sunday the mixing ratios are significantly smaller. Also the morning rushhour peak is shifted by about 2 hours on Saturday. Indicating that the observed NO_2 is mainly caused by traffic.

Cabauw (52°N, 5°E)

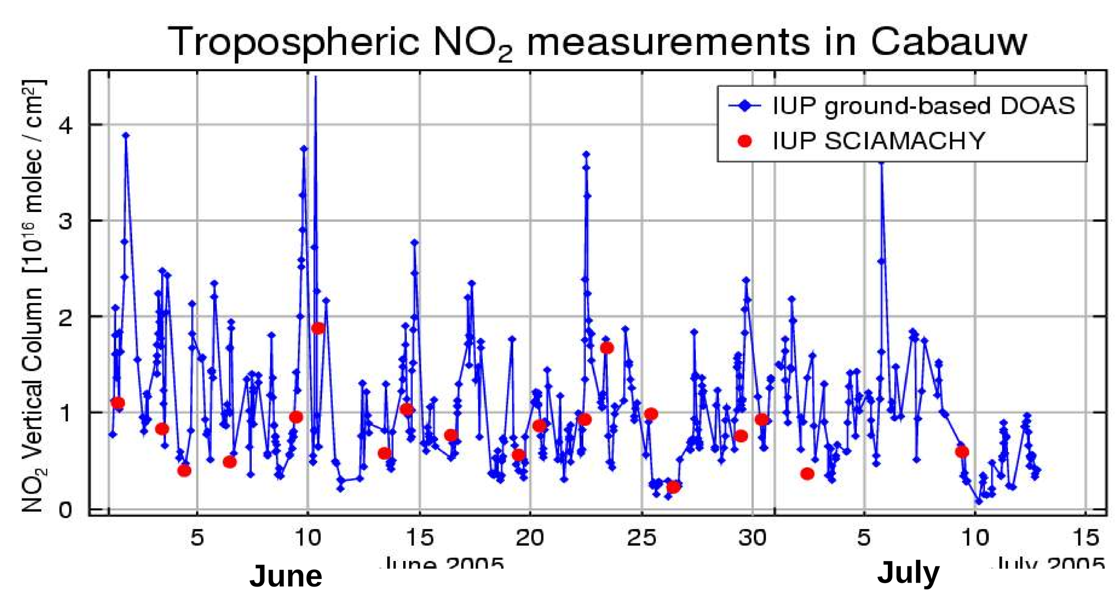


Figure 6 Tropospheric NO_2 columns from the MAX-DOAS compared to SCIAMACHY tropospheric columns.

From May to middle of July of this year a field campaign for OMI validation took place at Cabauw (The Netherlends). Various groups with different instruments participated including three MAX-DOAS set-ups.

The Figures 6 and 7 shows ground-based DOAS measurements compared to satellite data and to *in situ* measurements respectively. Excellent agreement is found for both tropospheric columns and NO_2 concentrations demonstrating the ability of the MAX-DOAS to work as a link between *in situ* and satellite observations.

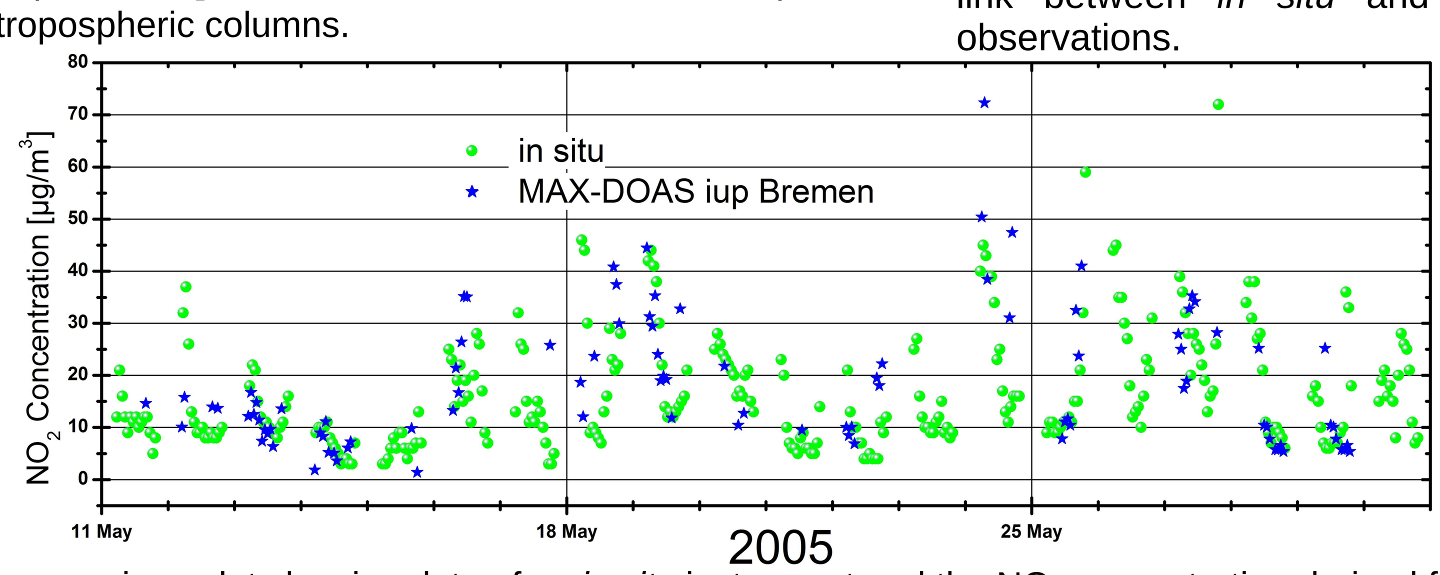


Figure 7 Intercomparison plot showing data of an *in situ* instrument and the NO_2 concentration derived from the lowest layer of 50 m thickness. Days with clouds are included.

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