

Estimation of NO₂ amounts emitted from the Portuguese wildfires in 2005: A synergistic use of observations by imaging and atmospheric instruments and chemistry-transport models

Julian Meyer-Arneke (1), Pieter Valks (2), Marianela Fader (1) and Thilo Erbertseder (1)

(1) German Aerospace Center DLR – DFD, Münchener Straße 20, 82234 Wessling, Germany

(2) German Aerospace Center DLR – IMF, Münchener Straße 20, 82234 Wessling, Germany

ABSTRACT

During August and September 2005, severe wildfires occurred on the Iberian Peninsula. Wildfires are generally associated with large emissions of NO₂ and other ozone precursors. In this study, tropospheric vertical NO₂ columns derived from SCIAMACHY observations are used to identify NO₂ emissions from wildfires and soils on the Iberian Peninsula. At the German Aerospace Center, tropospheric vertical NO₂ columns are operationally derived using a synergistic approach combining SCIAMACHY measurements and results from stratospheric and tropospheric chemistry models. First, the SCIAMACHY total slant NO₂ column is separated into a stratospheric and a tropospheric part. The stratospheric slant NO₂ column is derived from the chemistry and transport model ROSE/DLR. Secondly, the tropospheric vertical NO₂ column is computed from the tropospheric slant column by applying a tropospheric air mass factor (AMF). The AMF is calculated using the LIDORT radiative transfer model (v2.2+) and tropospheric NO₂ profile shape information derived from the EURAD/RIU air quality model. The EURAD model system simulates the physical, chemical and dynamical processes which control emission, production, transport and deposition of atmospheric trace species. The tropospheric NO₂ VCD derived with this method for the European area yield results which are qualitatively and quantitatively comparable with NO₂ column amounts derived from different approaches. Compared to the summer composite, the winter composite shows enhanced tropospheric NO₂ VCD due to increased anthropogenic emissions and different photochemistry. Enhanced tropospheric NO₂ VCD in the south western part of the Iberian Peninsula retrieved during August 2005 are to be attributed to the Portuguese wildfires. Enhancements of the tropospheric NO₂ VCD during September 2005 in the north western parts of the Iberian Peninsula are likely to be attributed to biogenic emission pulses following rain after a long dry period.

1 INTRODUCTION

Due to anthropogenic activity the chemical composition of the atmosphere is significantly influenced. Among all trace gases being emitted, oxides of nitrogen (in the following referred to as NO_x) play a central role. NO_x emission leads to hemisphere wide increase of tropospheric ozone, acidification and to an increased oxidation capacity in the troposphere. Moreover, NO_x has indirect effects on the Earth's radiative balance due to its influence on aerosol formation and on the burdens of many greenhouse gases like methane [1]. On the global scale, NO_x is mainly generated by combustion of fossil fuel or by biomass burning [2].

Soils have been identified to have a significant impact on the NO_x budget especially for unpolluted regions. In tropical regions, large pulses of biogenic NO_x emissions following rain on dry soils of savannas and seasonally dry forests are reported [3]. Long dry periods allow soils to accumulate inorganic nitrogen. The first rain then activates water-stressed bacteria leading to the consumption of the accumulated nitrogen and as a by-product to the release of NO pulses [3]. During the wet season, the NO_x emissions decrease but remain enhanced when compared to the dry season.

In this study, tropospheric NO₂ vertical column densities (VCD) are derived by combining SCIAMACHY measurements and stratospheric and tropospheric chemistry model results. In the following sections, the retrieval method is described, and tropospheric NO₂ maps for the European area are presented. The increase in tropospheric NO₂ due to the intense Portuguese wildfires in August 2005 are described, and the effect of biogenic soil emissions on the tropospheric NO₂ column over north-west Spain is discussed.

2 TROPOSPHERIC NO₂ COLUMN RETRIEVAL FROM SCIAMACHY

2.1 Methodology

The high spectral resolution of the space-borne spectrometer SCIAMACHY enables us to derive atmospheric column densities of NO₂ [4]. The core of the retrieval is a DOAS (Differential Optical Absorption Spectroscopy) fitting

technique that involves a multi-linear regression of measured optical densities against a number of reference spectra [5]. For the NO₂ retrieval, a fitting window between 425 and 450 nm is used.

Several methods to derive the nitrogen dioxide content in the troposphere are described in literature (e. g. [6], [7]). A key issue is the separation of the tropospheric and stratospheric contributions to the total NO₂ content. With the tropospheric excess method [6], the tropospheric and stratospheric contributions to the total NO₂ content are separated under the assumption of a relatively stable (both in time and space) NO₂ distribution. The NO₂ burden over the free Pacific Ocean determines the stratospheric amount as well as the tropospheric background. However, a constraint of this approach is the neglect of the stratospheric (zonal) variability. In order to consider this variability, we derive the stratospheric NO₂ contribution by means of the stratospheric chemistry transport model (CTM) ROSE/DLR, as described in section 2.2.

To account for the diurnal NO₂ cycle, the stratospheric analysis is calculated for exactly the overpass time of the satellite instrument. Using a geometric stratospheric air mass factor, the stratospheric NO₂ slant column density is derived from the model analyses. To avoid a bias, the analyses are scaled to “clean conditions” by means of a reference sector approach. In order to gain the tropospheric NO₂ slant column, the stratospheric slant column is subtracted from the total slant column as derived by the DOAS fitting. Similar approaches using CTM are used by [8] and [9]. The total NO₂ slant column is taken from the operational ESA product; currently SCIAMACHY Version 5.04. For further details on the total slant column retrieval the reader is referred to [10], [11]. The tropospheric NO₂ vertical column density (VCD) is derived from the NO₂ slant column by taking into account the surface reflectance, the viewing geometry and tropospheric NO₂ profile shape [12]. The latter is derived from the tropospheric air quality forecast system EURAD/RIU, as described in section 2.3. The deduction of tropospheric VCD from the NO₂ slant column is described in section 2.4.

2.2 Stratospheric CTM ROSE/DLR

The background distribution and the temporal evolution of stratospheric ozone and nitrogen dioxide are simulated using the 3D global chemical-transport model ROSE [13], [14]. The model covers the relevant gas-phase stratospheric chemical processes, and the heterogeneous processes on polar-stratospheric clouds and on sulfuric aerosols. It accounts for about 100 reactions, including oxygen, hydrogen, carbon, nitrogen, chlorine and bromine species. The chemical rate constants and cross-sections are taken from [15]. Photolysis rates are derived from a look-up table depending on solar zenith angle, ozone column and altitude. The chemical rate equations are solved by considering a chemical equilibrium state for the short-lived species (e.g. ClO, NO, HO, BrO). A semi-implicit scheme is used for the integration of the more stable reactants (e.g. HNO₃, N₂O, NH₄).

Wind and temperature fields are derived from 24-hour-forecasts performed using the global weather forecast model (GME) of the German Weather Service (DWD). By the means of optimal interpolation ozone columns derived from SCIAMACHY measurements are assimilated into the ROSE/DLR model (see also <http://wdc.dlr.de>).

2.3 Tropospheric model EURAD/RIU

The EURAD air quality forecast system consists of three major components: The PennState/NCAR mesoscale model MM5 to predict the needed meteorological variables, the EURAD Emission Module (EEM) to calculate the temporal and spatial distribution of the emission rates of the major pollutants, and the EURAD Chemistry Transport Model (EURAD-CTM) to predict the concentrations and deposition of the main atmospheric pollutants [16], [17].

The chemical mechanisms employed in the EURAD system are the so-called RADM2 and its successor RACM. They have been completed by the aerosol mechanism MADE (Modal Aerosol-Dynamics model for EURAD). The RADM2 mechanism contains 63 reactive species treated in 158 chemical reactions. Detailed aqueous phase chemistry for the treatment of the air pollutants is incorporated. The horizontal and vertical transport is done by using the 4th order Bott advection scheme. Vertical mixing of the species is treated by an implicit vertical diffusion scheme. The sink at the lower boundary of the model is treated by wet and dry deposition parameterization. The wet deposition relies on the predicted precipitation and the dry deposition is calculated via the deposition velocity for each species.

The EEM calculates the temporal and spatial distribution of the emission rates of the major pollutants from available data bases. The EEM processes different data bases ranging from continental scale down to local scale. The biogenic emissions are calculated online with respect to the given atmospheric condition (temperature, radiation, wind) and land type.

The forecast model system uses the method of nested simulations. This enables consistent modeling of air quality from small (local) to large (continental) scales. Therefore, the model resolution for the different domains varies between 1 to 125 km. We use the 125 km discretisation covering Europe. For all model domains a terrain following σ coordinate

with 23 unequally spaced vertical layers between the ground and 100 hPa is applied. Forecasted NO₂ profiles are collocated with SCIAMACHY observations in order to estimate the profile shape of the tropospheric NO₂ burden.

2.4 Derivation of tropospheric NO₂ vertical column density

The tropospheric NO₂ VCD can be calculated by dividing the tropospheric slant column by the tropospheric air mass factor. For an optically thin trace gas such as NO₂ at visible wavelengths, the air mass factor can be calculated using the altitude-resolved air mass factor approach [18]:

$$AMF = \frac{\int m(z) \cdot n(z) dz}{\int n(z) dz} \quad (1)$$

where $m(z)$ is the altitude-resolved air mass factor, $n(z)$ the NO₂ volume density profile in the troposphere and AMF the tropospheric air mass factor. In general, $m(z)$ depends on the altitude z , the viewing geometry, and the albedo and height of the reflecting surface (ground or cloud). However, $m(z)$ is independent of the NO₂ volume density profile $n(z)$. An altitude-resolved NO₂ air mass factor look-up table has been calculated with a spherical version of the multiple scattering radiative transfer model LIDORT [19], [20]. The NO₂ volume density profile is determined with the tropospheric air quality forecast system EURAD for each SCIAMACHY observation, as described above. Only SCIAMACHY observations with a cloud fraction smaller than 20% are used. Since even the smallest cloud fraction has a large effect on the air mass factor, cloud properties need to be taken into account. Here, we use SCIAMACHY cloud fractions as derived with the OCRA algorithm [21]. The air mass factor also strongly depends on the surface albedo, which is taken from a surface albedo climatology [22].

It is important to note, that the calculated tropospheric NO₂ VCD does not depend on the EURAD tropospheric NO₂ column, but only on the modelled NO₂ profile shape. While EURAD is best validated in the boundary layer, satellite observations are sensitive to the NO₂ in the mid and upper troposphere. So we expect the method to ideally combine both data sources.

3 TROPOSPHERIC NO₂ OVER EUROPE

The near-real-time computation of tropospheric NO₂ VCD has been performed since August 2005. This operational service is part of the GMES Service Element Project PROMOTE and is available at <http://www.gse-promote.org>. For this study, the period from August-October 2005 is considered to represent mean summer conditions, while the period November 2005 - February 2006 represents winter conditions (Fig. 2). In both cases, enhanced levels of pollution are seen over the heavily industrialised regions in the central and southern parts of the UK and the Benelux countries, as well as over major cities like Madrid, Paris, London and Moscow. Due to the short lifetime of NO_x in the atmosphere of about hours to days enhanced NO_x concentrations are strongly tied to their emission source. This indicates that fossil fuel combustion is likely to be the major source of NO_x over Europe. The tropospheric NO₂ columns are generally higher during wintertime. This observation is consistent with the fact that the amount of fossil fuel burnt during winter is higher than during summer due to cold weather conditions, and that the lifetime of NO₂ is longer during winter.

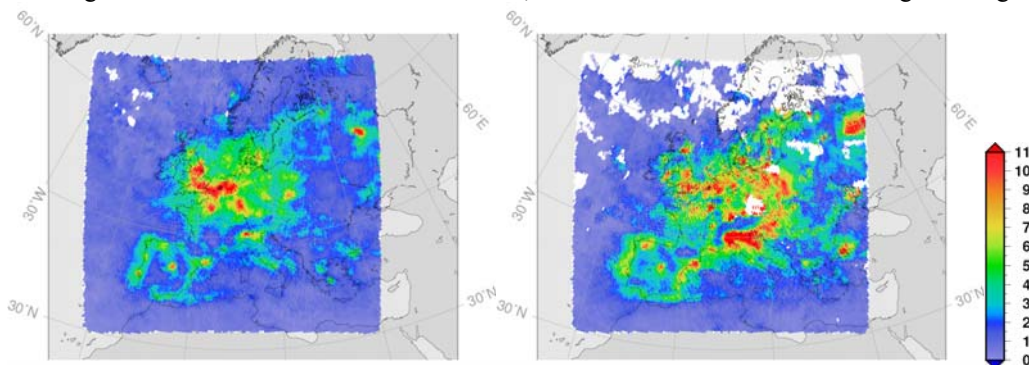


Figure 1: Summer (left) and winter (right) composite of tropospheric NO₂ vertical column densities (VCD) (given in 10¹⁵ molecules cm⁻²).

4 CASE STUDY: PORTUGUESE WILDFIRES AND BIOGENIC EMISSION

In August 2005 severe wildfires occurred on the Iberian Peninsula. Fig. 3 shows the hotspots indicating burning activity, as retrieved from AVHRR data [23]. In the beginning of August 2005, the number of detected hotspots increased fast, reaching its maximum around 11 August. Then the hotspot number decreased again, reaching its typical background value around 26 August. Trajectory calculations carried out using the trajectory model Traj.x [24] reveal that emissions from the Portuguese wildfires during August 2005 are transported into south westerly (onto the Atlantic Ocean) as well as into easterly directions. These fires were supported by dry conditions beginning in May 2005 and ending right at the beginning of October 2005. Fig. 4 (left) shows the volumetric soil water content (SWC) of the uppermost soil layer for the region Galicia derived from the operational analysis of the European Centre for Medium Range Weather Forecasts (ECMWF). After a major rainfall on 6 September, the SWC was significantly increased for about 10 days. It is very likely that this increase in soil humidity lead to biogenic emissions of nitric oxides, especially since Galicia is to be considered as a region with a comparably dense and active vegetation. Fig. 4 (right) shows the net primary production (NPP) derived from the BETHY/DLR vegetation model [25]. A trajectory analysis indicates that emissions were mainly transported northwards.

The difference between the tropospheric NO_2 fields in August and September 2005 is shown in Fig. 5. During August 2005, enhanced tropospheric NO_2 columns were found in the south western parts of the Iberian Peninsula, whereas during September, an increase of tropospheric NO_2 was observed in the northern parts of the Iberian Peninsula. The NO_2 increase during August can be attributed to the Portuguese wildfires, whereas the increase during September is due to biogenic emissions.

Trajectory analysis yield that the transport pattern of anthropogenic emissions from the densely populated cities Lisboa and Porto has been nearly equal during August and September 2005. Consequently, anthropogenic emissions do not contribute to the observed differences of the tropospheric NO_2 burden shown in Fig. 5.

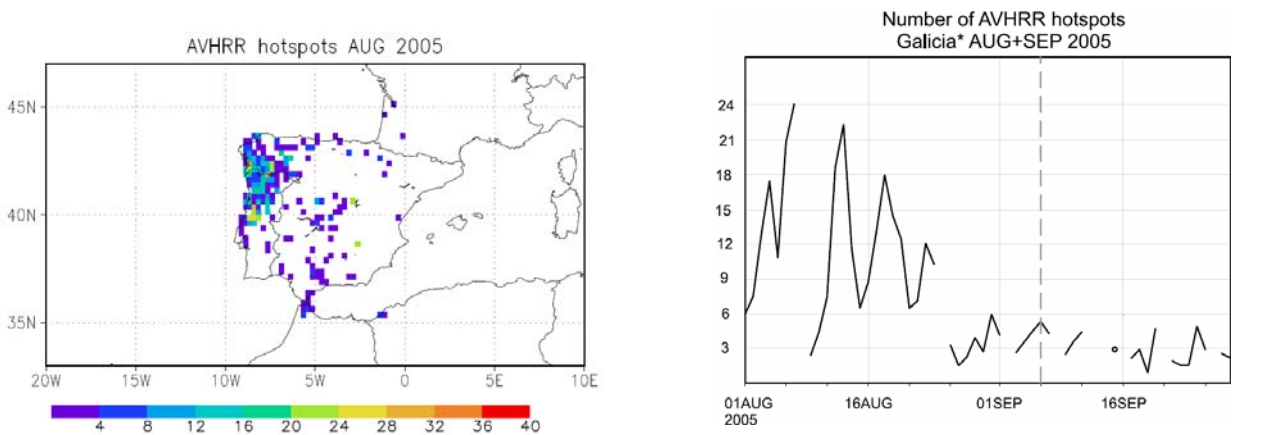


Figure 3: Number of hotspots derived from AVHRR for August 2005 (left). Number of AVHRR hotspots in the region from 9° W to 2° W and 40° N to 43,5°N (Galicia) (right).

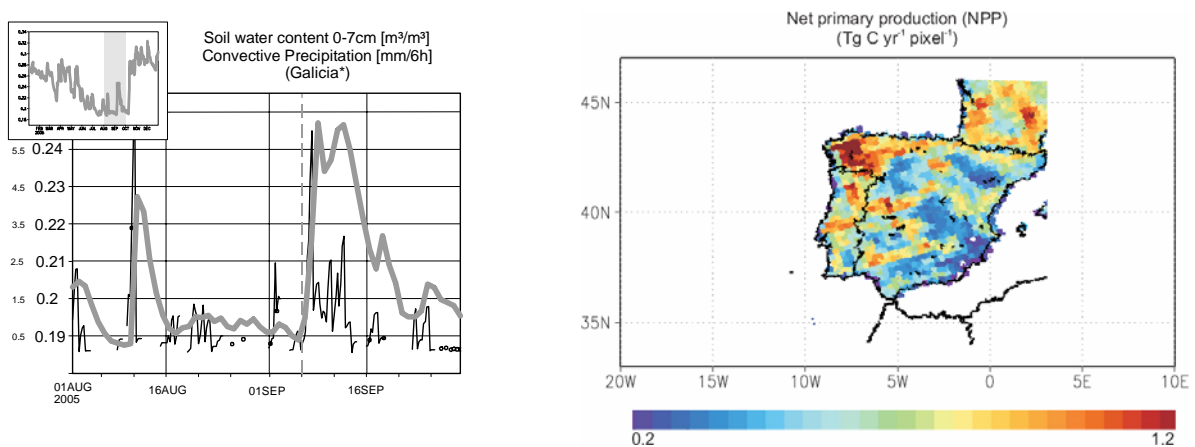


Figure 4: Net primary production (NPP) (left). Soil water content (grey) and convective precipitation (black) for Galicia (right).

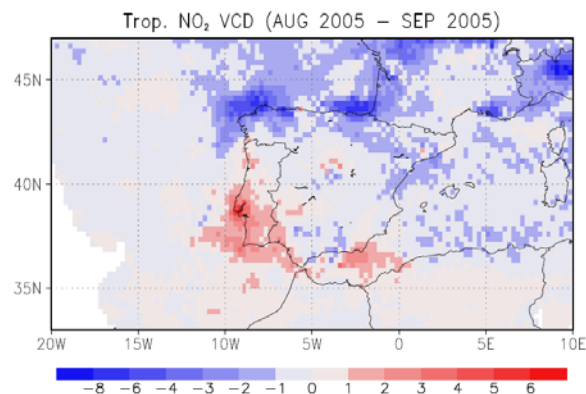


Figure 5: Difference between the August and September tropospheric NO₂ columns (given in 10¹⁵ molecules cm⁻²).

REFERENCES

- [1]: Prather, M. J., and D. Ehhalt (2001), Atmospheric chemistry and greenhouse gases, in *Climate Change 2001: The Science of Climate Change*, chap. 4, pp. 239–287, Cambridge Univ. Press, New York.
- [2]: FAO, Global estimates of gaseous emissions of NH₃, NO and N₂O from agricultural land, Food and Agricultural Organisation, Rome, 2001.
- [3]: Davidson, E. A. (1992), Pulses of nitric oxide and nitrous oxide flux following wetting of dry soil: An assessment of probable sources and importance relative to annual fluxes, *Ecol. Bull.*, 42, 149–155.
- [4]: Bovensmann, H., J. P. Burrows, M. Buchwitz, J. Frerick, S. Noël, V. V. Rozanov, K. V. Chance, and A. P. H. Goede: SCIAMACHY: Mission objectives and measurement modes, *J. Atmos. Sci.*, 56, 127-150, 1999.
- [5]: Platt, U., “Differential optical absorption spectroscopy (DOAS), *Air monitoring by Spectroscopic Techniques* (M. Sigrist, ed.)”, John Wiley & Sons, 1994, pp. 27–84.
- [6]: Richter, A., and J.P. Burrows, Retrieval of Tropospheric NO₂ from GOME Measurements, *Adv. Space Res.* 29, 1673-1683, 2002.
- [7]: Leue, C., M. Wenig, T. Wagner, O. Klimm, U. Platt and B. Jaehne, Quantitative analysis of NO_x emissions from GOME satellite image sequences, *J. Geophys. Res.*, 106, 5493-5505, 2001.
- [8]: Eskes, H. J. et al., GOME assimilated and validated Ozone and NO₂ fields for Scientific Users and Model Validation, Final, European Commission, Fifth Framework Programme, Environment and Sustainable Development, 1998-2002, April 2003.
- [9]: Richter, A., Burrows, J. P., Nüß, H., Granier, C, Niemeier, U., Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, 437, 129-132, doi: 10.1038/nature04092, 2005.
- [10]: DLR: ENVISAT SCIAMACHY Level 1b to 2 Off-line Processing Input / Output Data Definition Doc.No.: ENV-ID-DLR-SCI-2200-4, 2002.
- [11]: DLR, BIRA, SAO and ESA: Algorithm Theoretical Basis Document for GOME Total Column Densities of Ozone and Nitrogen Dioxide UPAS/GDOAS: GDP 4.0, Doc.No.: ERSE-DTEX-EOPG-TN-04-0007, 2004.
- [12]: Erbertseder, T., J. Meyer-Arnek, P. Valks and F. Baier, Derivation of Tropospheric NO₂ by Synergistic Use of Satellite Observations and Chemical Transport Modelling, in *Tropospheric Soundings from Space, ACCENT-TROPOSAT-2*, in 2004-5, page 223-228 .
- [13]: Rose, K. and Brasseur, G.: A three-dimensional model of chemically active trace species in the middle atmosphere during disturbed winter conditions. *J. Geophys. Res.* 94, 16 387–16 403, 1989.
- [14]: Baier, F., T. Erbertseder, O. Morgenstern, M. Bittner and G. Brasseur, Assimilation of MIPAS observations using a three-dimensional global chemistry-transport model, *Q. J. R. Meteorol. Soc.*, in print.
- [15]: Sander, S. P., A. R. Ravishankara, R. R. Friedl, W. B. De-More, D. M. Golden, C. E. Kolb, M. J. Kurylo, M. J. Molina, R. F. Hampson, R. E. Huie, and G. K. Moortga: Chemical kinetics and photochemical data for use in stratospheric modeling. Eval. 13, JPL Publ., 00–3., 2000
- [16]: Jakobs, H.J., H. Feldmann, H. Hass, and M. Memmesheimer: The use of nested models for air pollution studies: an application of the EURAD model to a SANA episode. *J. Appl. Meteor.*, Vol. 34, No. 6, 1301-1319, 1995.

- [17]: Jakobs, H.J., E. Friese, M. Memmesheimer, A. Ebel: A real-time forecast system for air pollution concentrations. Proceedings of EUROTRAC Symposium 2002, 2002.
- [18]: Palmer, P. I., D. J. Jacob, K. Chance, R. V. Martin, R. J. D. Spurr, T. Kurosu, I. Bey, R. Yantosca, A. Fiore, and Q. Li, Air-mass factor formulation for differential optical absorption spectroscopy measurements from satellites and application to formaldehyde retrievals from GOME, *J. Geophys. Res.*, 106, 17,147-17,160, 2001.
- [19]: Spurr, R. J. D., T. P. Kurosu, and K. Chance: A linearized discrete ordinate radiative transfer model for atmospheric remote sensing retrieval, *J. Quant. Spectrosc. Radiat. Trans.*, 68, 689-735, 2001.
- [20]: Spurr, R. J. D., Simultaneous derivation of intensities and weighting functions in a general pseudo-spherical discrete ordinate radiative transfer treatment, *J. Quant. Spectrosc. Radiat. Transfer*, 75, 129-175, 2002.
- [21]: Loyola, D., Cloud Retrieval for SCIAMACHY, ERS-ENVISAT Symposium, Gothenburg, 2000.
- [22]: Koелеmeijer, R. B. A., J. F. de Haan, and P. Stammes, A database of spectral surface reflectivity in the range 335 – 772 nm derived from 5.5 years of GOME observations, *J. Geophys. Res.*, 108(D2), 4070, doi:10.1029/2002JD002429, 2003.
- [23]: Gesell, G., German Remote Sensing Data Center (DFD), German Aerospace Center (DLR), personal communication, 2006.
- [24]: Meyer-Arneke, J., A. Ladstätter-Weißmayer, A. Richter, F. Wittrock, and J. P. Burrows: A study of the trace gas columns of O₃, NO₂ and HCHO over Africa in September 1997, *Faraday Discussions*, DOI: 10.1039/b502106p, 2005.
- [25]: Knorr, W. 1997. Satellite Remote Sensing and Modelling of the Global CO₂ Exchange of Land Vegetation: A Synthesis Study (Satellitengestützte Fernerkundung und Modellierung des globalen CO₂-Austauschs der Landbiosphäre: Eine Synthese). Max-Planck-Institut für Meteorologie, Examensarbeit Nr. 49, Hamburg, Germany. ISSN 0938-5177.