Algorithm Description

SCIAMACHY NO_2 Tropospheric Columns

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1 Forward Model:

The tropospheric NO_2 Column product uses airmass factors derived with the radiative transfer model SCIATRAN (Rozanov et al., 1997), This model includes the effects of multiple scattering and atmospheric refraction and sphericity, but not polarisation.
The data analysis which is based on the DOAS technique (see next section) uses Lambert Beer's law of absorption and thus implies the "forward model" of an optically thin atmosphere.

2 Inversion Procedure:

The SCIAMACHY tropospheric NO_2 analysis is performed in a series of steps, each of which will be described in more detail below. A more detailed description of the data analysis for GOME can be found in Richter and Burrows, 2002. The SCIAMACHY NO2 retrieval is described in Richter et al., 2004, Richter et al., 2005 and Nuess, 2005.
Briefly, the first step is the retrieval of the total amount of NO_2 along the effective line of sight (Slant Column SC). This is determined using the well known Differential Optical Absorption Spectroscopy (DOAS) technique (e.g. Platt, 1994). The slant column depends among others on the viewing geometry, the solar zenith angle and the amount and vertical distribution of the absorber in the atmosphere. The second step of the analysis is the correction for stratospheric absorption which is achieved by subtracting the NO2 column over a clean reference region. The third step is the conversion of the remaining tropospheric slant column to a geometry independent tropospheric vertical column using an airmass factor (AMF) based on radiative transfer calculations. As the airmass factor is strongly dependent on a priory assumptions, a large effort went into improving the reliability of the input used.
A simplified formula describing the overall method used to derive the vertical tropospheric columns is

$$VC_{trop} = \frac{SC - SC_{strat}}{AMF_{trop}} = \frac{SC - SC_{ref}}{AMF_{trop}}$$

where SC is the slant column, VC the vertical column, AMF the airmass factor and the index strat and trop stand for the stratosphere and troposphere, respectively and ref denotes the measurements taken on the same day at the same latitude over the reference sector.

2.1 DOAS Fit

The Differential Optical Absorption Spectroscopy (DOAS) method is based on the optical absorption spectroscopy but deals only with those parts of the absorption that vary rapidly with wavelength. As in standard absorption spectroscopy, the optical depth is derived by taking the natural logarithm of the ratio of the measured intensity and a background measurement without absorption. In the case of the SCIAMACHY measurements described here, the first is a nadir earth-shine measurement I and the latter is a measurement of the solar irradiance I_0 which is taken once per day.
The optical depth observed is determined by absorption by trace species in the atmosphere, and scattering by molecules, aerosols and clouds, all of which follow Beer's law. As the nadir measurements are scattered light measurements, scattering is not only relevant for the extinction, but at the same time together with surface reflection the light source used. In general, the wavelength dependence of scattering is λ^{-4} for Rayleigh scattering and $\lambda^{-1..1.5}$ for Mie scattering, and can be approximated by a polynomial in wavelength. The equation used for the DOAS analysis thus can be written as

$$\ln\frac{I(\lambda)}{I_0(\lambda)} = -\int_0^\infty \sum_i \rho_i(s)\sigma_i(s,\lambda)ds - \sum_j c_j\lambda^j$$

where σ_i is the absorption cross-sections of species i, ρ_i its density, λ the wavelength and the integral is taken along the light path. The c_j are coefficients determined by a fit, and are used as closure parameters. As this polynomial in wavelength also compensates any slow changing parts of the absorption, only differential structures in the absorption cross-sections can be used (thus the D in DOAS).
Assuming that the absorption cross sections do not depend on height (small pressure and temperature dependencies), σ_i can be removed from the integral and the slant column, which is the total amount of absorber integrated along the light path can be defined:

$$SC_i = \int_0^\infty \rho_i(s)ds$$

Using this definition, the DOAS equation now is

$$\ln\frac{I(\lambda)}{I_0(\lambda)} = -\sum_i SC_i\sigma_i(\lambda) - \sum_j c_j\lambda^j$$

With I and I_0 measured by the instrument and σ_i known from lab measurements, the searched quantities SC_i can be determined by a linear fit from the measurements together with the coefficients c_j. Details on the implementation of the DOAS retrieval can be found in Richter, 1997. Details on the settings used can be found in Table 1:

Parameter	Value
wavelength window	425 – 450 nm
absorption cross-sections	NO_2 (Bogumil et al., 2003, 243K) O_4 (Greenblatt et al.(modified), 1990) H_2O (Rothmann et al., 1992) Ring (Vountas et al., 1998)
empirical functions used	none
degree of polynomial	quadradic
offset and slope correction	offfset
background spectrum	daily ASM solar measurement
normalisation	none
data source	uncalibrated lv0 and lv1 data

Table 1: DOAS settings used for the NO_2 retrieval from SCIAMACHY measurements

2.2 Correction of the stratospheric contribution

In the sunlit stratosphere, NO_2 columns between $0.5x10^{15}$ and $8x10^{15}$ molec/cm^2 are found depending on latitude and season. This stratospheric NO_2 column contributes strongly to the total NO_2 signal measured by SCIAMACHY, and thus must be subtracted from the derived columns to isolate the tropospheric column. As the sensitivity of SCIAMACHY to NO_2 in the stratosphere and the troposphere is different, the correction for the stratospheric contribution is best done on the level of slant columns.

As a result of the sun-synchronous orbit of ENVISAT, the SCIAMACHY overpass is at the same local time at all longitudes for one latitude, and therefore also at the same solar zenith angle. Stratospheric NO_2 columns depend largely on solar zenith angle and day length, and therefore are in first approximation zonaly invariant if sampled at the same local time. Thus, SCIAMACHY measurements taken at the same latitude and on the same day over a clean region of the Earth can serve as an estimate of the stratospheric NO_2 slant column for all longitudes. This approach is used in the "reference sector method", where the Pacific sector (180° - 210°) is used as clean air background.

While this method provides a reasonable correction of the stratosphere in most cases, it has two drawbacks: first, any inhomogeneities in the stratospheric NO_2 fields will introduce errors in the tropospheric NO_2 products, and second the small but non-zero tropospheric NO_2 column over the Pacific is forced to zero and the numbers retrieved constitute not the total tropospheric column but rather the tropospheric excess column with respect to the selected clean air region.

For GOME data, a more sophisticated correction of the stratospheric NO_2 column has been developed based on SLIMCAT simulations, but this approach has so far not been implemented for SCIAMACHY. A discussion of other possible approaches to separate tropospheric and stratospheric NO_2 can be found in *Richter et al., 2002*.

2.3 Airmass Factors

The tropospheric slant columns depend on many factors such as viewing geometry, solar position, vertical distribution of the absorber, surface elevation, surface albedo, aerosol optical depth, aerosol type and aerosol vertical distribution. Clouds also have a large impact on the sensitivity of the measurements, but are not yet fully treated in the retrieval.

To convert the measured slant columns in vertical columns which are independent of all the above quantities, appropriate airmass factors have to be computed with a radiative transfer model. Here, the SCIATRAN model (*Rozanov et al., 1997*) is used which was formerly known as GOMETRAN. While AMFs can easily be computed by SCIATRAN with high accuracy, it is not obvious what to use for the input parameters such as absorber profile and albedo, and thus the error budget of the tropospheric NO_2 columns is dominated by the uncertainties of the AMFs.

2.3.1 Vertical Absorber Profile

The sensitivity of nadir measurements in the UV/visible spectral domain decreases strongly towards the surface. To compensate for this, the vertical distribution of the absorber must be known, an information which can not be derived from the SCIAMACHY measurements themselves. It is important to realize, that only the shape of the profile needs be known, whereas the absolute amount has no impact on the airmass factor within a very large concentration range.

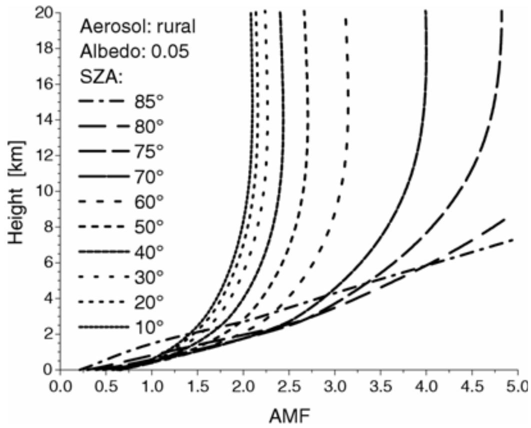

Fig. 1: Dependence of AMF on SZA for a surface albedo of 0.05 and rural aerosol

The only possible source of global estimates for vertical absorber profiles are tropospheric chemical transport models, that based on emissions, transport and chemistry predict trace gas concentrations at a number of altitude levels on global grids.
However, the use of model profiles in the airmass factor calculation raises a number of questions, mainly related to the fact, that a priori assumption is introduced into the SCIAMACHY data product which then can no longer be viewed as an independent data source:

- In regions with large emissions in the model, the profile will have its maximum close to the surface, and the corresponding AMF will be small. If the emissions used in the model are wrong, and in fact there is no surface peak in NO_2, then the SCIAMACHY NO_2 values are enhanced and tend to reproduce the model even though there is no enhanced NO_2 in the raw measurements.
- Similarly, in regions where the model underestimates the emissions, the SCIAMACHY NO_2 will also underestimate the real values.
- As a result of the relatively coarse grid of the model, the profiles constitute average values and will tend to "smear out" local plumes in the satellite data.
- The emission scenarios used so far do not account for diurnal or seasonal variations, and thus might introduce biases in the data.

All these problems must be kept in mind, in particular when comparing measurements and model results. Probably the only way to deal with them is to always use profiles from the respective model when intercomparing data.
Notwithstanding these uncertainties, using model profiles will increase the overall accuracy of the tropospheric GOME columns significantly, in particular on average. Therefore, daily MOZART (*Horowitz et al., 2003*) model profiles have been used for the AMF calculation.

2.3.2 Surface Elevation

Surface elevation plays a role in airmass factor calculation as the height dependence of the sensitivity makes GOME more sensitive to NO_2 from say Mexico City than from a city on sea level. Thus, the surface elevation is taken into account in the AMF calculations.

2.3.3 Surface Albedo

The sensitivity of GOME to the lower troposphere depends strongly on the surface albedo, at least in situations where the aerosol optical depth is not too large. Therefore, a good estimate is needed for surface albedo at the wavelengths used for the retrieval (425 - 450 nm) as a function of location and season. Such a data base has been derived from GOME data by

Koelemeijer et al., 2003. This data set is used in the AMF calculations for the SCIAMACHY NO_2 data.

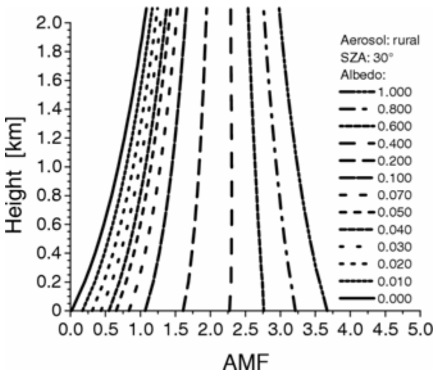

Fig. 2: Dependence of AMF on surface albedo for a rural aerosol and 30° SZA

As in the case of model profiles, the use of an albedo climatology will improve the accuracy of the tropospheric NO_2 columns on average. However, for individual measurements quite large errors can be introduced if the actual surface albedo differs from the one in the climatology as a result of snow coverage, land use change or variations in albedo below the resolution of the data base (1° x 1°).

2.3.4 Aerosols

The effect of aerosols on the tropospheric NO_2 columns measured by GOME is twofold: NO_2 within or below an aerosol layer will be seen with reduced sensitivity. NO_2 situated above a reflecting aerosol will be seen with enhanced sensitivity. It therefore is necessary to know the vertical distribution of aerosols and also the composition, as absorbing and reflecting aerosols will have different effects on the radiative transfer.

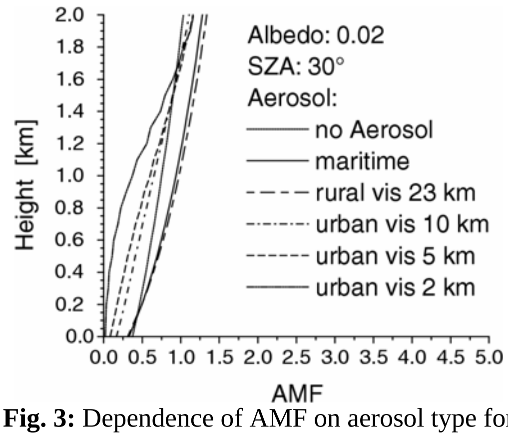

Fig. 3: Dependence of AMF on aerosol type for an albedo of 0.02 and a SZA of 30°

Unfortunately, there is very little information available on the distribution of aerosols, and the large variability in space and time makes this input particularly difficult to assess. Therefore, for the current SCIAMACHY data products a very simple approach was selected based on the aerosol parameterisation of the LOWTRAN model (*Shettle and Fenn, 1976*). Three types of aerosols were distinguished: a maritime aerosol (23km visibility, 70% humidity) over the oceans, an urban aerosol with 10 km visibility and 70% humidity over industrialised regions and a rural aerosol (23km visibility, 70% humidity) elsewhere.

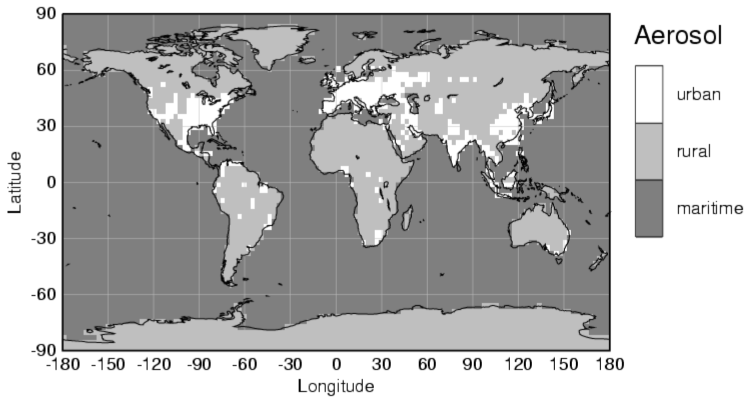

Fig. 4: Assumed distribution of aerosol types

This simplified treatment of aerosols introduced significant errors into the NO_2 data product. in particular, NO_2 in biomass burning regions and in very polluted regions such as China will be underestimated. Sensitivity studies show, that for an urban aerosol with 2 km visibility the sensitivity of SCIAMACHY to NO_2 in the lowest km is close to zero. Also, NO_2 might be overestimated on clear days in the industrialized regions. However, without more detailed information on the actual aerosol profiles, not much can be done to improve the results.

2.3.5 Clouds

The effect of clouds on the NO_2 retrieval is similar to that of aerosols, only much stronger: clouds effectively shield the atmosphere below them from view and strongly enhance the sensitivity to any absorption above them, in particular close to the cloud top. There also is some enhancement in sensitivity to absorption within the upper part of the cloud.
Two different approaches can be used to account for the impact of clouds: either a simple cloud screening algorithm can be used to select only data below a certain cloud threshold or the impact of clouds can be modelled in detail based on measurements of cloud fraction and cloud top height and assumptions on the amount of NO_2 below the cloud. For the SCIAMACHY NO_2 columns, a simple threshold technique was used based on the integrated intensity of the measurement itself. When compared to FRESCO cloud cover, it corresponds to a maximum cloud cover of about 20%, a rather large value chosen to avoid large data gaps in the Northern Hemisphere in winter and spring. Clearly, there is a risk of underestimation of NO_2 by using this generous threshold, but detailed analysis of the data shows that there is only a weak dependence of NO_2 slant column on cloud cover in winter over Europe and North America. This is probably the result of NO_2 above low clouds, and will reduce the errors introduced. This is probably not true for regions such as India during the Monsoon season, where persistent cloud cover prevents GOME from measuring down to the surface over weeks at a time.

2.4 Implementation

The implementation of the retrieval algorithm is based on the block airmass factor concept. Airmass factors are computed for 100 m thick layers in the atmosphere for different parameters (SZA, albedo, aerosol types, surface albedo, elevation) for all altitudes between the surface and 20 km and saved in lookup tables. For every measurement, the elevation, aerosol type and surface albedo values are retrieved from the data bases, and the SZA from the measurement time and location. Using these inputs, the appropriate block AMF are retrieved from the data base and multiplied with the appropriate tropospheric absorber profile from the MOZART run:

$$AMF = \frac{\sum VC_i^{MOZ} AMF_i}{\sum VC_i}$$

where the VC_i^{MOZ} are the partial columns in the individual layers of the MOZART profile and the AMF_i the airmass factors for the different layers.
This parameterisation relies on the fact that the atmosphere is optically thin at the wavelengths of interest, an assumption that is valid in most situations. It is computationally fast and conceptually very similar to the weighting function approaches used by other groups.

Parameter	Source	Values	Interpolation
Albedo	*Koelemeijer et al.*	0.00, 0.01, 0.02, 0.03, 0.04, 0.05, 0.07, 0.10, 0.20, 0.40, 0.60, 0.80, 1.00	linear
SZA		10, 20, 30, 40, 50, 60, 70, 75, 80, 85	linear
Surface Height	TerrainBase Global DTM Version 1.0	0 - 9000m, 100 m layers	nearest neighbour
Aerosol	LOWTRAN	2.815° x 2.815°, urban 10km, rural 23km, maritime 23 km	nearest neighbour
Vertical Profile	MOZART	MOZART sigma levels, interpolated on AMF grid (100 m layers)	Akima interpolation
Latitude / Longitude		2.815° x 2.815°	nearest neighbour

Table 2: Sampling points and interpolation schemes used in the AMF calculation

3 Auxiliary Data:

The auxiliary data used for the airmass factor calculations has already been noted above (surface albedo, aerosol loading, surface height, vertical NO_2 distribution).

4 Sensitivity and Error Analysis and Algorithm Validation:

Tropospheric SCIAMACHY NO_2 columns have not yet been rigorously been validated:

- Results of two validation campaigns are given below, but they are restricted to relatively polluted situations in two northern hemisphere mid-latitude locations.
- Validation with airborne AMAXDOAS measurements is reported in Heue et al., 2005
- The data have also been compared to Brewer NO_2 measurements at NASA-Goddard as reported in Cede et al., 2006.

However, a similar data product has been produced using GOME measurements, and this data set has been compared to independent measurements in a number of studies (Heland et al., 2002, Petritoli, 2004, Schaub et al., 2005, Irie et al., 2005, Ordonez et al., 2006).

4.1 Comparison with GOME measurements

Thus, comparison of SCIAMACHY and GOME measurements for the same time period can give an indication of the consistency of the SCIAMACHY data set. As shown in Fig. 4, the agreement between the two data sets is good if the GOME data are selected to have the same gaps as the SCIAMACHY measurements and only the low resolution backscans of SCIAMACHY are used.

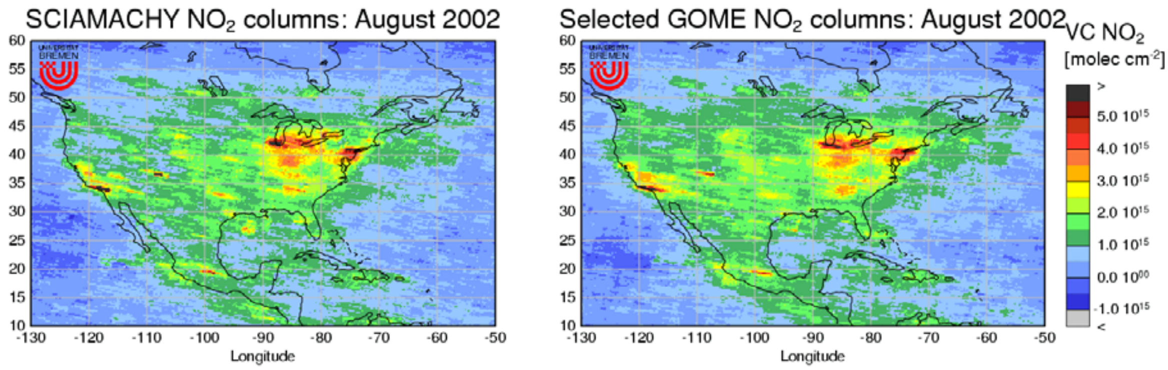

Fig. 5: Comparison of GOME (right) and SCIAMACHY NO_2 tropospheric vertical columns for August 2002. Only the larger backscan has been used for SCIAMACHY and only GOME measurements for which a corresponding SCIAMACHY pixel exists have been included.

4.2 Validation with ground-based MAXDOAS measurements

During the DANDELIONS campaign in June 2005, the IUP Bremen performed MAXDOAS measurements at Cabauw in the Netherlands. This site was particularly well suited for validation as it is characterized by the absence of large local sources but at the same time often experiences large tropospheric NO2 columns as a result of transport from cities and industry in the surrounding. The MAXDOAS measurements can directly provide a measurement of the tropospheric column with little dependence on the vertical distribution of the NO_2, making these data ideal for validation of satellite columns.

In Fig.6, the results of a comparison between ground-based and SCIAMACHY columns is shown. A good correlation of 0.72 was found with a slope of 0.87 (+-0.2) that in this case indicates an overestimation by the satellite measurements. In this comparison, it turned out to be crucial to both use the right time of measurement for the MAXDOAS data (they were interpolated to the time of satellite overpass) and to limit the comparison to the closest SCIAMACHY pixels. In this comparison, only the closest pixel within a 50 km radius was used. Relaxing the radius to 100 km already reduces the correlation substantially.

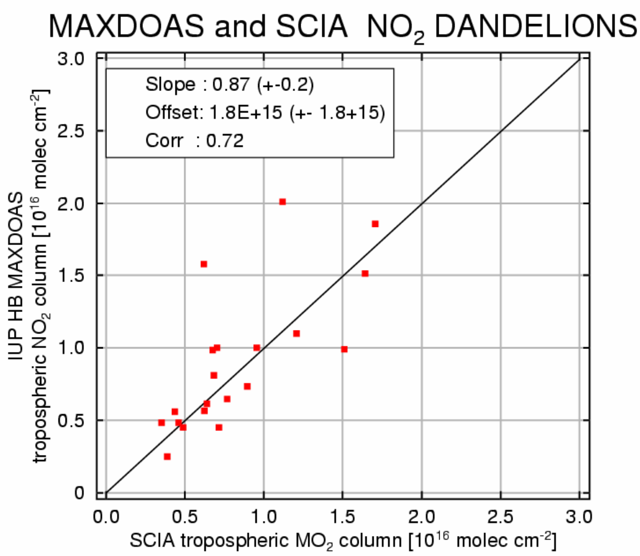

Fig. 6: Comparison of SCIAMACHY (x-axis) and ground-based (y-axis) NO_2 tropospheric vertical columns for measurements taken during the DANDELIONS campaign.

The tropospheric NO_2 retrieval has a number of problems, mainly related to the airmass factors and cloud treatment which impact on the data quality:

- in the presence of clouds, the column is usually underestimated but can also be overestimated if some NO_2 is above the clouds
- the vertical profiles in the atmosphere will usually deviate from the model climatology used, introducing large errors for individual measurements
- varying aerosol loadings can greatly affect the columns derived
- as the current data product uses a simple reference sector method for the correction of the stratospheric component, measurements in high latitudes in winter and spring are potentially biased if the stratospheric NO_2 distribution is not symmetric
- also, as the columns are only "tropospheric excess columns", they are too low by the amount of NO_2 found in the troposphere over the reference region (180° – 210°)

5 Recommendations for Product Validation:

Validation of tropospheric NO_2 columns should be dome in as homogeneous situations as possible using only clear sky data. The time of measurement is critical as is the distance between validation measurement and satellite overpass. In the case of air-borne measurements, the extrapolation to the surface and above flight altitude has a large impact on the results and needs to be performed carefully.
As aerosols have the largest impact in winter at mid and high latitudes when the sensitivity to the surface layer is reduced, the accuracy of the tropospheric NO_2 columns is expected to be lower at these times.

6 Data Availability

Tropospheric NO2 columns from SCIAMACHY measurements are available in different forms:

1. Image archive at
   http://www.iup.physik.uni-bremen.de/doas/scia_data_browser.htm
2. ASCII files with monthly averages on a 0.125° x 0.125° grid at
   http://www.iup.physik.uni-bremen.de/doas/scia_no2_data_tropos.htm#V07
   Thee files are password protected; the password can be obtained from Andreas Richter.
3. ASCII files with individual orbit swath data are available on request from Andreas Richter.

Images are available without constrains as long as they are properly referenced. Data files are provided under the conditions that

- they are not passed on without permission
- we are kept informed on relevant results coming from the use of the data
- should the data form a substantial part of a publication, we are asked to be co-authors

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