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Supplement of

Cloud and aerosol classification for 2.5 years of MAX-DOAS observations in Wuxi (China) and comparison to independent data sets

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Supplement:

Analysis of the O₄ absorption

The analysis of the O₄ dSCDs is performed in the spectral range of 350 nm - 391 nm covering two O₄ absorption bands using the DOAS method (Platt and Stutz, 2008). For the analysis the WINDOAS software (Fayt and van Roozendaal, 2009) was used. Apart from the O₄ cross section at 296 K (Greenblatt et al., 1990), also cross sections of NO₂ at 294 K (Vandaele et al., 1998) and O₃ at 293 K (Bogumil, 2003) were included. Effects of broad band scattering and extinction were accounted for by including a third order polynomial. The effect of rotational Raman scattering is considered by including a Ring spectrum (Grainger and Ring, 1962; Solomon et al., 1987; Wagner et al., 2009) computed by the DOASIS software (Kraus, 2006). Also an intensity offset is included in the fitting routine. All spectra are allowed to be shifted and stretched against a Fraunhofer reference spectrum (FRS). Fig. S1 shows typical DOAS fit results for the O₄ analysis. It should be noted that we discard 69 measurement sequences, for which the root-mean-square (RMS) deviation of the residual structure larger than 3×10^{-2} or shifts larger than ± 1 detector pixels.

The O₄ concentration is proportional to the square of the oxygen concentration (Greenblatt et al., 1990) and thus varies only slightly with time (due to variations of temperature and pressure). Thus for the conversion to AMF (see equations 3 and 4 in the main text) we used a constant VCD_{O₄} of 1.25×10^{43} molecules² cm⁻⁵ (for the unit see Greenblatt et al. (1990)). This value is calculated from average values of the surface temperature (290K) and surface pressure (1010 hPa) at Wuxi. Here it should be noted that during the course of one year we found deviations of the O₄ VCD from the selected value of ± 10 %. Since in this study we use the O₄ measurements only for the identification of fog (see main text), these deviations don't have an effect on the cloud primary classification results. Also the effect of the fog classification is very small.

For the interpretation of the O₄ AMF, another important aspect has to be considered: to remove the strong solar Fraunhofer lines, a FRS is included in a DOAS

fit. Since the FRS used in our analysis also contains atmospheric O₄ absorption, the result of the DOAS fit actually represents the difference between the O₄ SCDs of the measured spectrum and the FRS. This difference is usually referred to as differential SCDs (dSCD). Like the O₄ SCDs (eq. 3 in the main text), also the O₄ dSCD can be converted into the corresponding O₄ dAMF (eq. 4 in the main text).

We investigated the effect of the instrumental degradation on the O₄ dSCDs. We find systematic temporal variations of O₄ dSCD (see Fig. S2a) that are probably caused by variations of the spectral resolution of the instrument (see Fig. S2b). Because of the systematic variations we did not use the O₄ absorption measured in zenith for the identification of optically thick clouds. Instead we identify optically thick clouds by largely reduced values of the measured radiances.

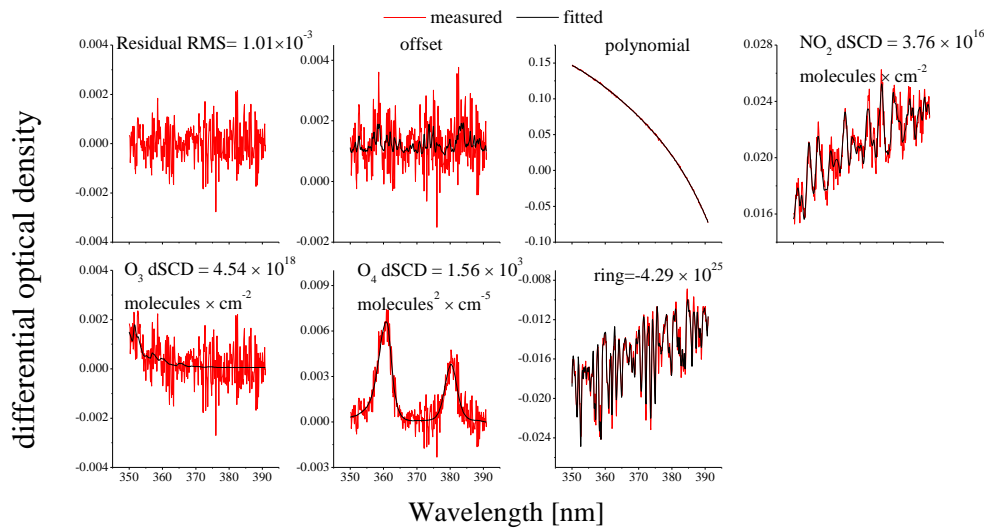


Fig. S1 Example of a DOAS fit for a spectrum measured at an elevation angle of 5° at 11:47 on September 15, 2012 using the Fraunhofer reference spectrum from the same elevation sequence. The red lines indicate the trace gas cross sections scaled to the corresponding absorptions in the measured spectrum; the black lines indicate the respective fit results. The residual (top left) indicates the difference between the measured spectrum and all fitted spectra.

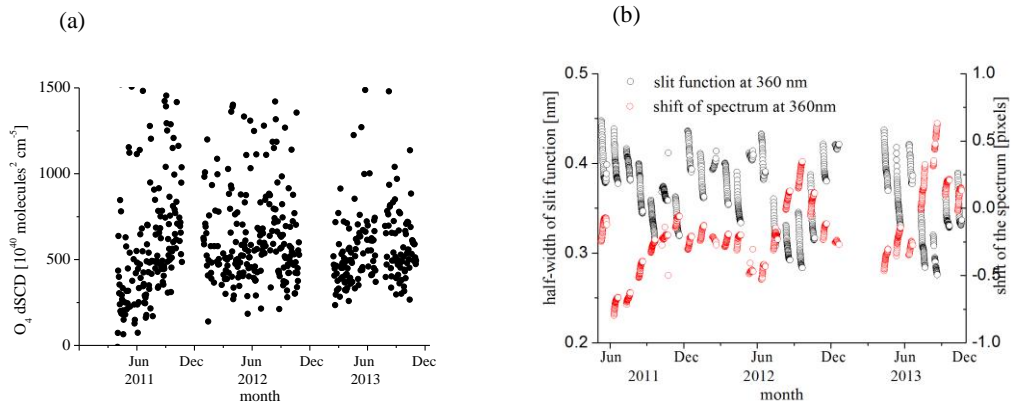


Fig. S2 (a): O₄ dSCD for zenith view in the SZA range of 49° to 51° from the retrievals with a fixed reference spectrum (from Sep 15, 2012). (b): The full width at half maximum and spectral shift (red points) of the measured spectra derived from a fit to a high-resolution solar spectrum.

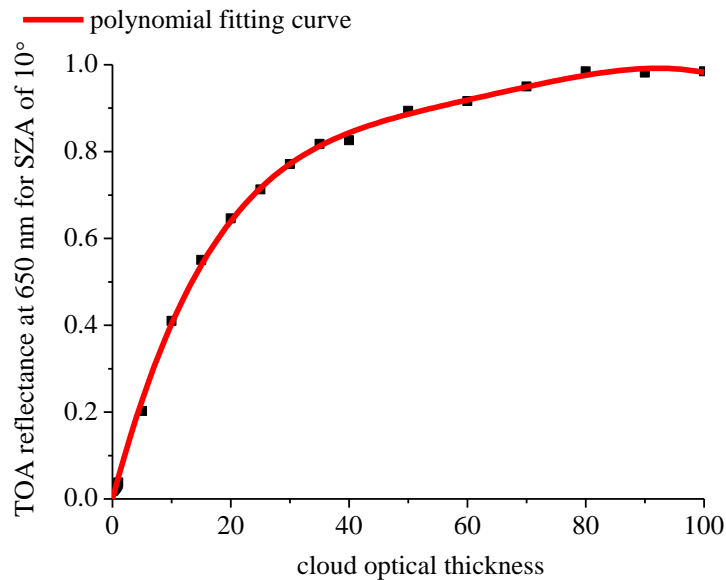


Fig. S3 Top of the atmosphere (TOA) reflectance at 650 nm for nadir view for SZA of 10° as a function of the cloud optical thickness derived from radiative transfer simulations. The cloud optical properties are described by the Henyey Greenstein approximation with an asymmetry parameter of 0.85 and a single scattering albedo of 1. The cloud layer range is from 1 to 2 km for COT of 1 to 15, from 1 to 3 km for COT of 20 to 30, from 1 to 4 km for COT of 35 to 40, and from 1 to 5km for COT of 50 to 100.

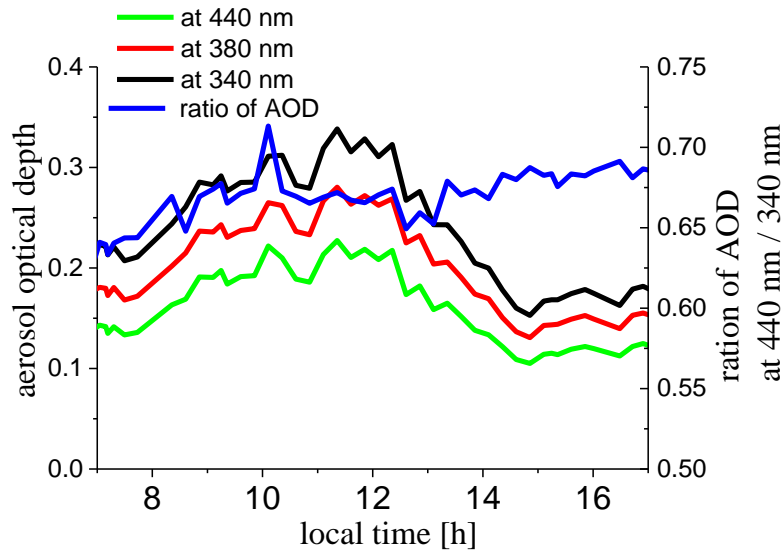


Fig. S4 Time series of the AOD at 340 nm (black), 380 nm (red) and 440 nm (green) on 29 July 2012 as well as the ratio of the AOD at 440 and 340 nm (black) (right axis) in the sky condition of “extremely high midday CI”.

Comparison of cloud products from MODIS, OMI and GOME-2

We briefly discuss the satellite cloud products in more detail and compare them with each other. Here we make use of the fact that the overpass time of MODIS on Terra is similar to that of GOME-2, and the overpass time of MODIS on Aqua is similar to that of OMI to compare the cloud products from GOME-2 and OMI with MODIS.

In Fig. S5a the effective CFs from OMI are compared with the corresponding MODIS results (on Aqua). Because the effective CF is clipped between 0 and 1, we excluded the data with effective CF of 100% and smaller than 1% for this comparison. A good agreement with the fitted linear slope of 0.71, the intercept of 6.7% and the square of correlation coefficient (R^2) of 0.61 is found. The different spatial resolution and slightly different overpass time of the two instruments contribute to the scattering of the points. The different retrieval techniques may also contribute to the discrepancy of effective CFs between both satellite instruments. In Fig. S5b in the same way the effective CFs from GOME-2 are compared with the corresponding MODIS results (on Terra). A good agreement with the fitted linear

slope of 0.86, the intercept of 14% and the R^2 of 0.61 is found. The larger effective CF from GOME-2 than that from MODIS is found, especially when the COT is low. This phenomenon has been found by Acarreta et al. (2004). Besides different spatial resolution and overpass time, the different spatial collocation criteria of GOME-2 and MODIS also contribute to the discrepancy between them.

The retrieval algorithms using the O_4 absorption for OMI and the O_2 (FRESCO+) for GOME-2 normally retrieve much larger CP than MODIS, especially for optically thick clouds. The reason is that multiple scattering leads to the fact that the cloud pressure from the O_4 absorption algorithm and FRESCO+ is closer to the middle of the clouds than to the top of the clouds (Acarreta et al., 2004; Wang et al., 2008; Wang and Stammes, 2014). The comparisons of the CPs from OMI and GOME-2 with those from MODIS in Fig. S5c and d also show the same feature.

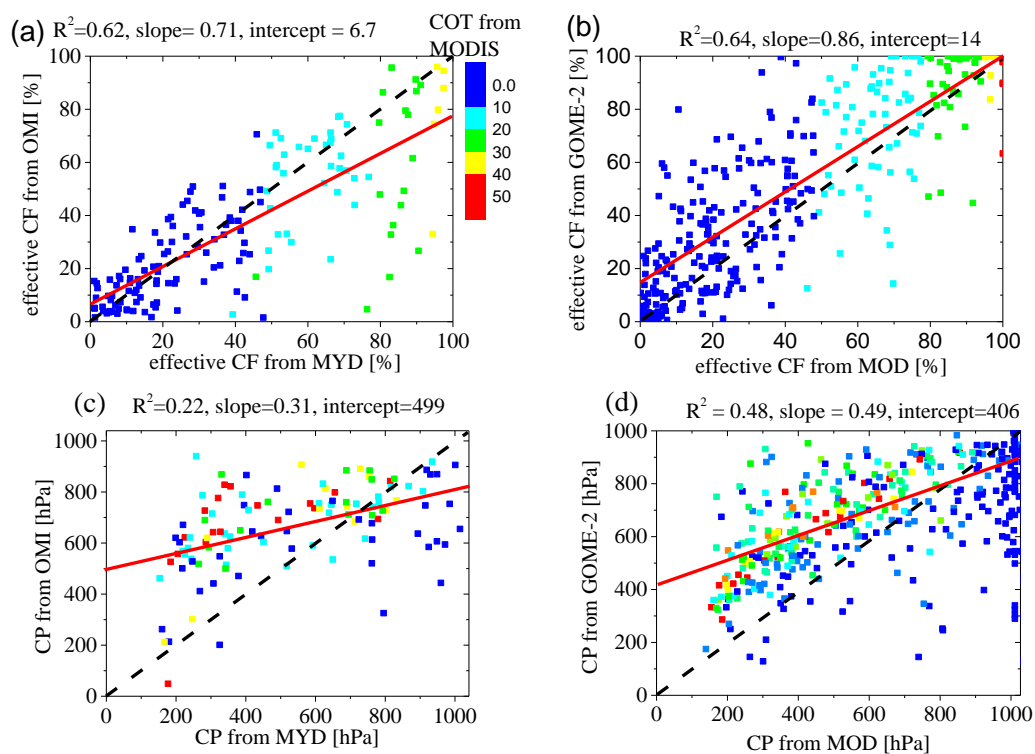


Fig. S5 Comparison of cloud products derived from GOME-2 or OMI and MODIS: effective CF from OMI and GOME-2 are plotted against those from MODIS on Aqua (MYD) a) and on Terra (MOD) b), respectively; CPs from OMI and GOME-2 are plotted against those from MODIS on Aqua (MYD) c) and on Terra (MOD) d), respectively. The black dashed line indicates the 1:1 line. The colours indicate the COT from MODIS. The comparisons are performed for satellite measurements over the Wuxi site. For the comparison of CPs, the data with corresponding effective CF <

20% are excluded to only keep the cloudy conditions, because the CP can't be accurately retrieved for effective CF < 20%.

Selection of threshold values

The procedure for the selection of threshold values is described. Firstly the days with specific sky conditions were selected based on visual images from MODIS (Fig. S6), time series of AOD at 340 nm from Aeronet (Fig. S7a), and the visibility at 550 nm derived from the visibility meter (Fig. S7b):

-24 September 2012: clear sky with low aerosol

-29 September 2012: clear sky with high aerosol

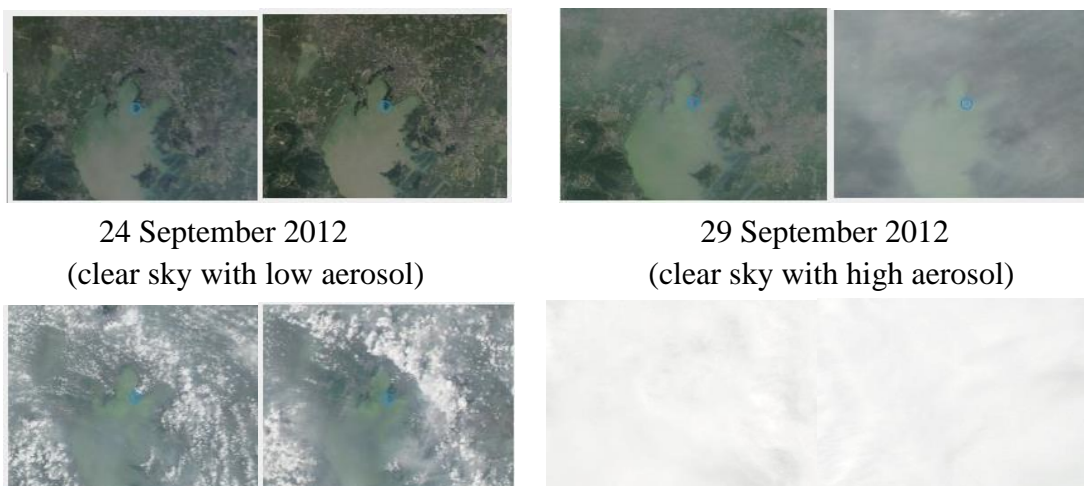
-20 July 2012: broken clouds

-9 September 2012: continuous and thick clouds (the COT from MODIS is around 50)

-28 July 2012: cloud holes

-10 June 2012: fog

Fig. S8 depicts the time series of the quantities described in Table 1 for the six selected days. From visible inspection of the different quantities for the different sky conditions on the selected days we determined the respective threshold values, which are listed in Table 1. In Fig. S9 exemplary results of the cloud classification scheme are presented for the selected days (including also the days shown in Figs. 7 and 8 of the main manuscript). Overall the deduced sky conditions are consistent with those identified by the MODIS images, and the measurements from AERONET and the visibility meter.



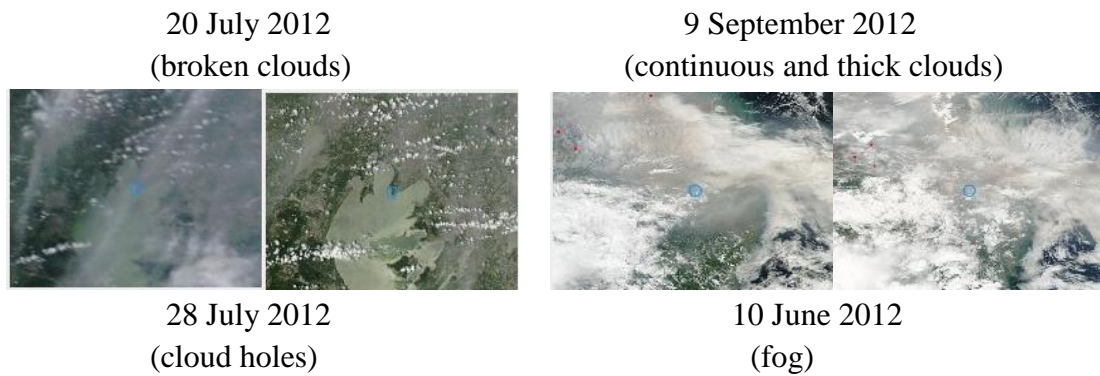


Fig. S6 Visual images from MODIS (left Terra; right: Aqua) for the six selected days with different sky conditions near Wuxi.

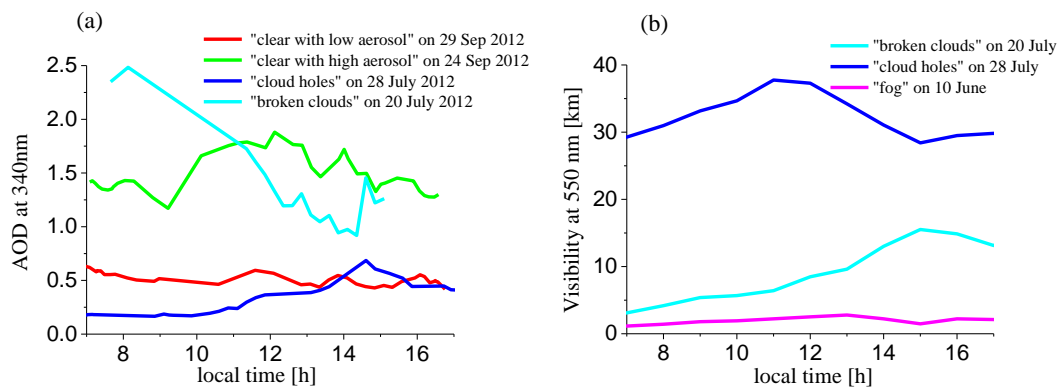
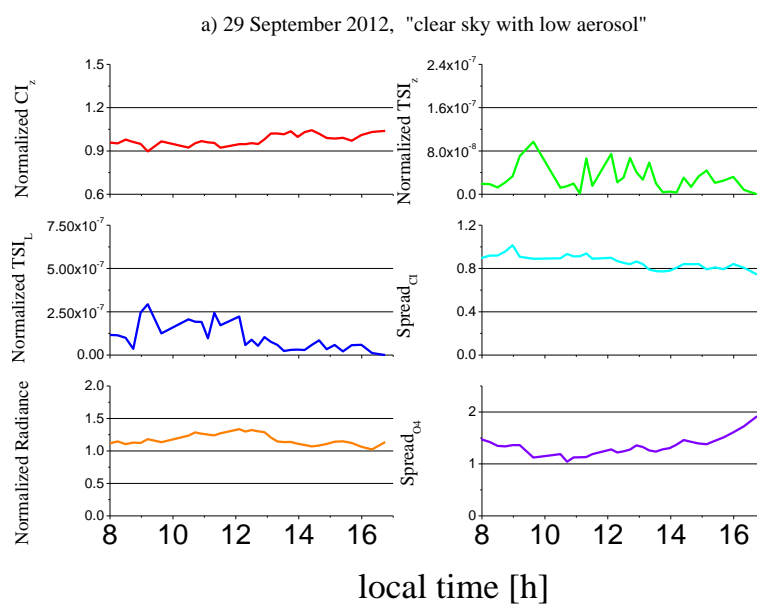
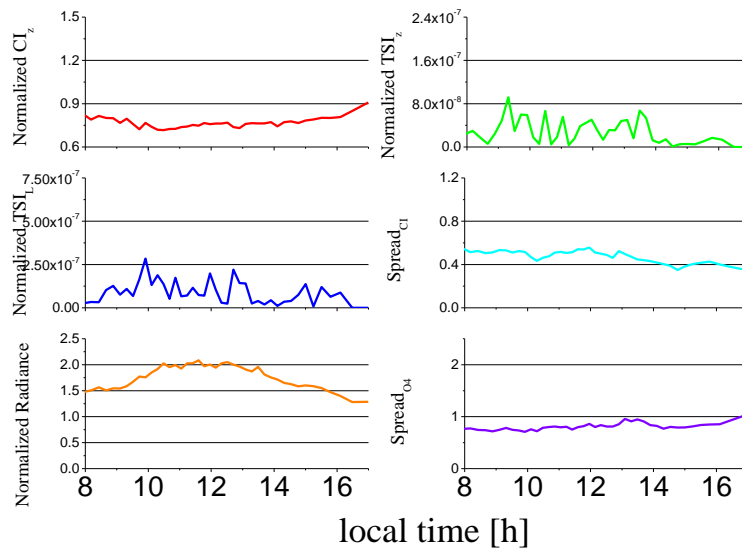


Fig. S7 (a) Time series of the AOD at 340 nm from AERONET on the days with clear sky, cloud holes or broken clouds (AOD data are not available for the other days because of clouds); (b) Time series of the visibility at 550 nm from the visibility meter on the days with broken clouds, cloud holes and fog (visibility data are not available for the other days due to instrumental problems).

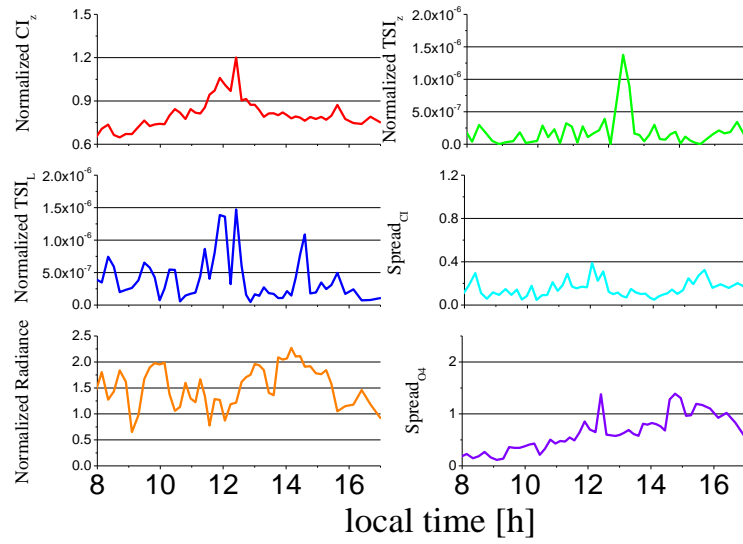


local time [h]

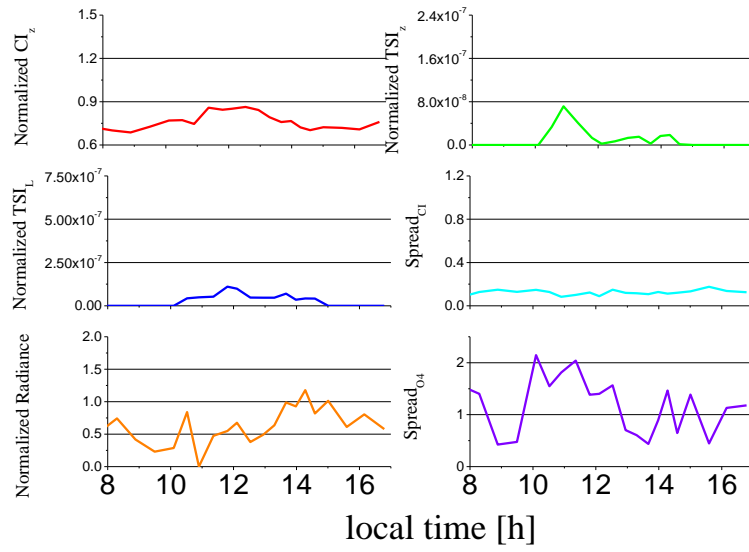
b) 24 September 2012, "clear sky with high aerosol"



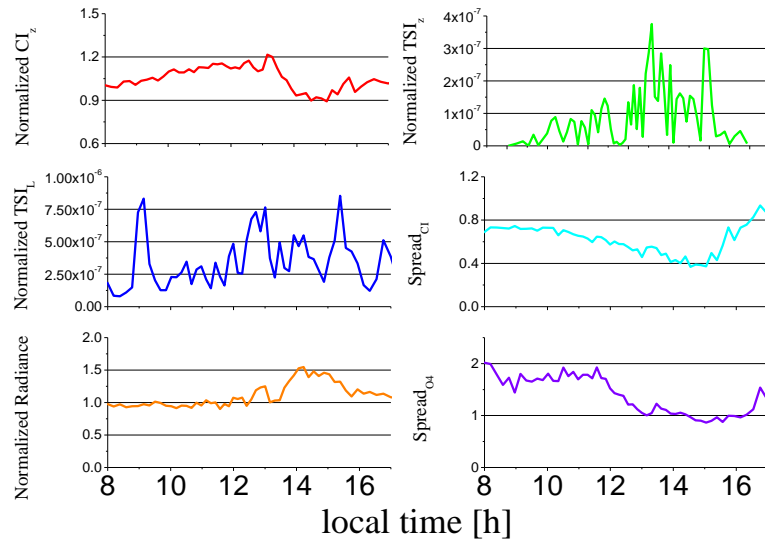
c) 20 July 2012, "broken clouds"



d) 9 September 2012, "continuous clouds"



e) 28 July 2012, "cloud holes"



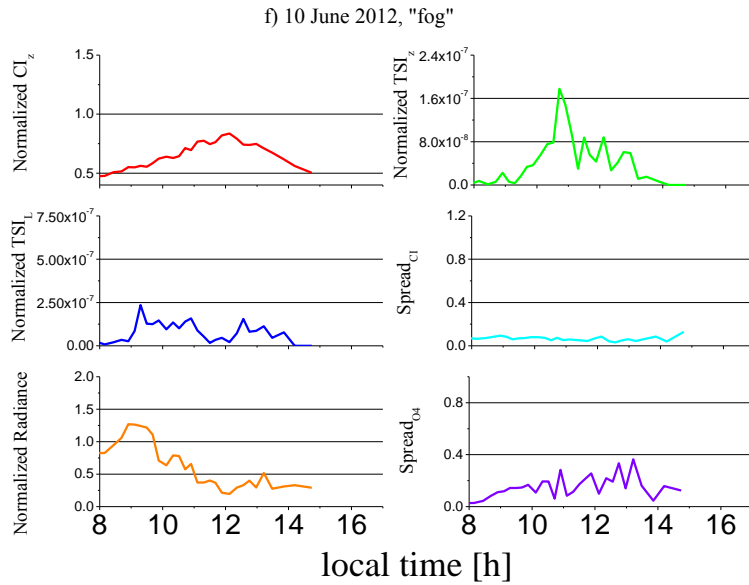


Fig. S8 Time series of the normalized CI_z , normalized TSI_z , normalized TSI_L , spread $_{CI}$, normalized radiance and spread $_{O4}$ on the six selected days with the typical different sky conditions.

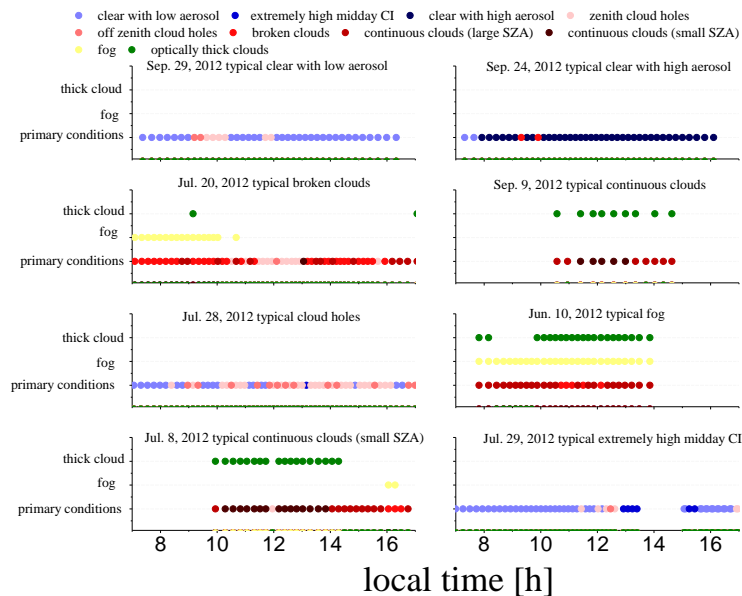


Fig. S9 Results of the cloud classification scheme for the selected days. Each symbol represents an individual elevation sequence. In the lower part of the individual figures, the primary sky conditions are indicated. In both upper parts the secondary sky conditions 'optically thick clouds' and 'fog' are indicated.

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