## **Supplementary Information**

## The CU 2D-MAX-DOAS instrument - part 2: Raman Scattering Probability Measurements and Retrieval of Aerosol Optical Properties

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Fig S1. Sensitivity studies of the dRSP fit coefficient as a function of the spectral fit window at visible wavelengths. The example is taken from 22 July 2012 at 8:18 LS (EA = 3°, AA = 0°). The top row shows the RMS, and the bottom row shows the dRSP, calculated as the relative difference in percent  $\left[\frac{(dRSP - dRSP_0)}{dRSP_0}\right]x100\%$ , where  $dRSP_0$  is the value obtained with the standard settings (black circle), and dRSP is the value obtained changing the lower, upper limit of the wavelength window, and polynomial order. For comparison with the standard settings an X is shown in the other windows. The theoretical RMS calculated based on photon counting statistics is 0.0013.



Fig S2. Same as Figure S1, but for evaluation windows at UV wavelengths. The graphs represent multiple analysis of the same spectrum also shown in Fig. S1. As can be seen the retrieval of dRSP in the UV shows more variability and higher RMS in comparison with the visible. The theoretical RMS calculated based on photon counting statistics is 0.0035. The open circle represents the standard setting following Wagner et al. (2009b).



Fig S3. Same as Fig.4 (main text) but for a  $SZA = 35^{\circ}$ .



Figure S4. Effect of the AOD in the RSP (left) and  $R_{norm}$  (middle) as a function of SRAA for four SZAs (rows). For clarity, the RSP as a function of AOD for the same SZAs is shown in the right column (similar to Fig. 7 in the main text). Additional parameters are g = 0.64, SSA = 0.98, SA = 0.05.



Fig S5. Comparison between the dRSP retrieved using a direct-sun (x-axis) and zenith sky (y-axis) spectrum as reference spectrum in the DOAS analysis. All spectra recorded with the solar almucantar scan on 22 July 2012 (SZA  $< 75^{\circ}$ ) are shown. The dRSP values are color coded by the absolute SRAA. The intercept is negative because of the higher RSP contained in the zenith sky direction. The direct sun reference spectrum has a lower, but non-zero RSP (see main text for details).



Fig S6. Asymmetry Factor Parameter ( $AFP_{I_{norm}}$ ) in percentage (color code) on (A) 17 July and (B) 22 July 2012. The radii of the polar plot represents the local time, the angles are the absolute SRAA (see main text for details).