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Interactive comment on “Influence of clouds on the oxidising capacity of the troposphere” by L. K. Whalley et al.

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The authors used measured $j(\text{O}^1\text{D})$ photolysis frequencies as a model input. The $j(\text{O}^1\text{D})$ measurements were made with a filter radiometer on a 22 m tower and were covering downward radiation from the upper hemisphere. In fact, because of low ground albedos upward radiation from vegetated surfaces can usually be neglected in the UV-B range, at least for ground based measurements. The same applies for the tower measurements at Mt. Schmücke in good approximation. However, if the tower is situated into a cloud, upward radiation will increase dependent on cloud optical thickness and tower height. In a very thick cloud the radiation field can become virtually isotropic with up-welling radiation as strong as down-welling. Because of the limited

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tower height this was probably not the case here, but nevertheless $j(\text{O}^1\text{D})$ could have been significantly enhanced.

To estimate the potential contribution of upward radiation, simulations with the TUV model (also used by the authors) were consulted for the Mt. Schmücke station on 01 Oct 2010 (mid of the campaign period). Spectral actinic flux densities were calculated assuming a range of solar zenith angles (SZA), a typical ozone column of 300 DU, standard aerosol, a ground albedo of 0.02, and an elevation of 937 m. Model output was generated for 959 m representing the tower top at 22 m above ground. Moreover, a homogeneous cloud cover of 1000 m thickness was assumed starting directly at the ground and extending to about 2 km cloud top elevation which is typical for continental stratus clouds. The total cloud optical depth (COD) was varied and from the simulated spectra photolysis frequencies $j(\text{O}^1\text{D})$ were calculated.

In a first step the ratios of downward $j(\text{O}^1\text{D})$ under overcast and clear sky conditions was calculated as a function of COD as shown in Fig. 1. These calculations reveal a non-linear dependence that can be utilized to estimate the COD encountered during the campaign: a reduction by 70% as found experimentally corresponds to a COD of about 30-40. These CODs are in reasonable agreement with those that can be estimated from the liquid water content (LWC) measured at the tower top (Petty, 2006):

$$\text{COD} \approx \frac{3L}{2\rho_l r_{\text{eff}}} \quad (1)$$

Her L ist the liquid water path, ($L = \text{LWC} \times 1000 \text{ m}$), ρ_l is the density of liquid water and r_{eff} is the effective cloud droplet radius which was assumed to be $10 \mu\text{m}$. LWCs between 0.1 and 0.3 g m^{-3} that were measured at the tower result in CODs between 15 and 45.

In a second step the ratio total/downward $j(\text{O}^1\text{D})$ was calculated as shown in Fig. 2. Here a nonlinear increase is observed. For the COD estimated above the enhancement factor is about 1.2. Consequently, when the tower is in clouds the measured $j(\text{O}^1\text{D})$

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should be scaled up accordingly. The same applies for $j(\text{HCHO})$

References

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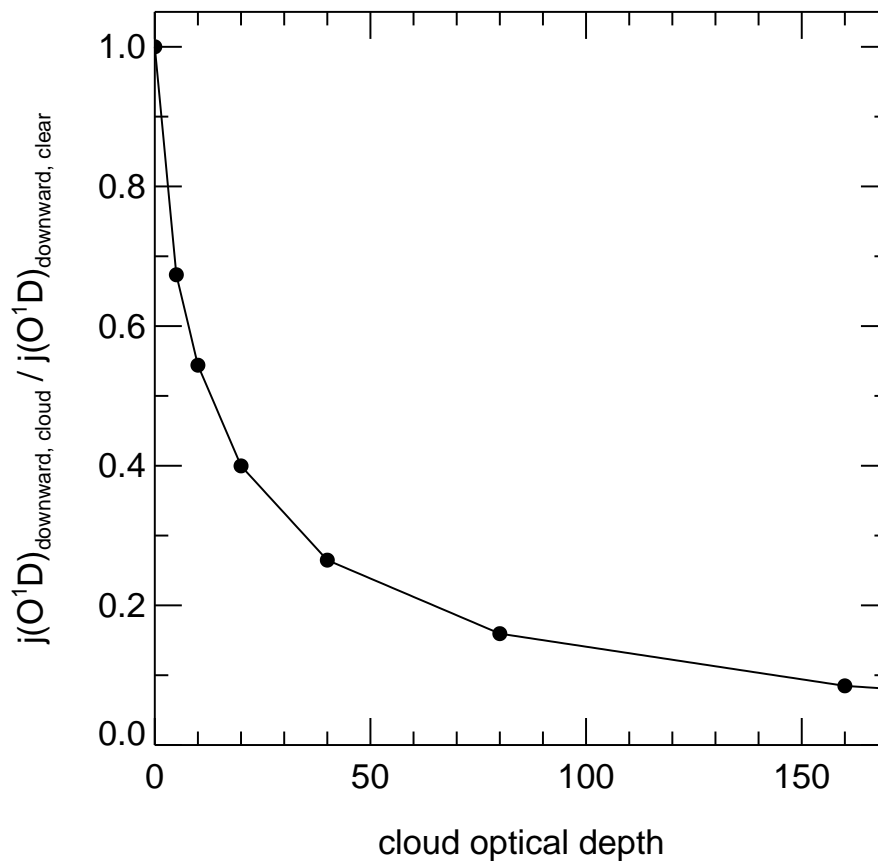
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Fig. 1. TUV simulated COD-dependent ratios of downward photolysis frequencies under overcast and clear sky conditions on 01 Oct 2010. The ratios exhibit little dependence on SZA.

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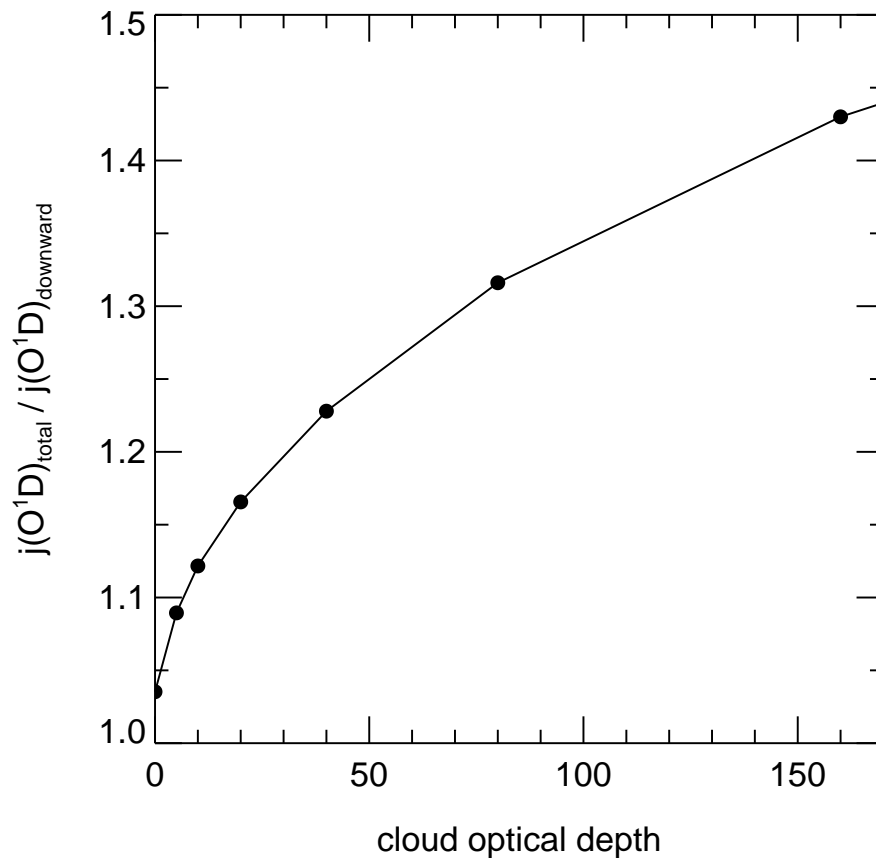
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Fig. 2. TUV simulated COD-dependent ratios of total and downward photolysis frequencies on 01 Oct 2010. The ratios exhibit little dependence on SZA.

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