Response to J.-F. Müller's interactive comment on the manuscript "Modeling global impacts of heterogeneous loss of HO$_2$ on cloud droplets, ice particles and aerosols"

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We thank Dr. J.-F. Müller for his critical and insightful comments related to our manuscript. This critique has motivated us to examine our modeling assumptions once again. Upon inspection of the changes in the HO$_2$ mixing ratios due to heterogeneous loss on cloud droplets we feel that , indeed, our assumption of instantaneous mixing within a grid-cell, still leads to an over-estimation of the effect of cloud-uptake for grid-cell average values for instances when cloud cover (CC) is low. An improved assessment, as detailed below, suggests that in our current manuscript we over estimate the cloud effect on HO$_2$ loss on a global scale by approximately a factor 2-3. When accounting for this, a first estimate suggests that combined liquid and ice cloud uptake is no longer the dominating sink for HO$_2$ when compared to other sink terms on aerosol, but rather of equal magnitude. A scaling factor to the loss of HO$_2$ on cloud, which accounts for sub-grid scale (SGS) mixing, can subsequently be introduced to provide an updated evaluation of HO$_2$ uptake on all respective reactive surfaces.

For this purpose, we have reconstructed a HO$_2$ concentration field that is a best estimate of the assumption of no-mixing within a grid-cell. This assumption is likely more realistic, considering the relatively large mixing time scale between cloudy and non-cloudy air. The reconstruction is done by combining two independent 1-day simulations with the C-IFS, where HO$_2$ concentrations are modeled to be representative either for within the cloud ($HO_{2,cloudy}$) or outside the cloud ($HO_{2,free}$). To obtain an estimate for $HO_{2,cloudy}$, the reaction rate representative for in-cloud HO$_2$ loss is applied without any scaling to the whole grid-cell. Next, the two resulting instantaneous HO$_2$ concentration fields are scaled with *CC* to obtain a grid-cell average concentration, according to Eq. 1, which could serve as a best-estimate of grid cell average HO$_2$ in the situation of no-mixing.

$$HO_{2,no\ mix} = (1 - CC)HO_{2,free} + CC\ HO_{2,cloudy} \qquad\qquad (1)$$

This reconstructed HO$_2$ field contrasts to the 'instantaneous mixing' (IM) approach as followed in our present manuscript, where reaction rates are scaled with cloud fraction, rather than the resulting HO$_2$ concentrations. Resulting mean HO$_2$ mixing ratios of the various approaches for a single day (1 April 2008), given as a function of CC, are presented in Fig 1. For practical reasons a no-mixing approach (NM) i.e. where calculations are performed both for in-cloud and cloud-free chemistry separately within each grid cell, is difficult to achieve in a CTM.

With the availability of the reconstructed HO$_2$ field we can now quantify the estimated error of the IM approach with respect to NM. The net loss of HO$_2$ at 800hPa for IM is over-estimated by, on average, ~35% for grid cells with CC>0.01. This lower limit for CC is also applied in the C-IFS for activating cloud chemistry and scattering in the photolysis routine. When averaging over all grid cells containing the full range of CC values, the difference between the two methods is ~17%.
Fig. 1 illustrates that the mean HO$_2$ mixing ratios in IM still follows the same general shape as the reconstructed HO$_2$, representative for NM. For high CC (> 0.8) the resident HO$_2$ mixing ratios become fairly similar between approaches, where both are more reasonable than when assuming no HO$_2$ uptake on cloud at all. However, the frequency distribution of CC at global scale (not shown) reveals a large contribution with CC < 0.1. Specifically, over ~50% of all grid cells with CC>0.01 have a value below 0.1. In summary, the change in HO$_2$ at low CC contributes most to the global average, which is where the IM approach makes the largest error.

Also when comparing the average decrease in daily mean HO_2 mixing ratios with respect to the case of no HO_2 cloud uptake at all, this is ~21% at 800hPa for IM (as applied in run AER-CLD in our manuscript) and ~7% for NM. This again is largely defined by the contributions of grid-cells with low CC, which occur mostly in the tropics. Note also that here photochemical activity, and thus HO_2 mixing ratios, are higher than in the extra-tropics. Figure 2 shows a zonal mean in the HO_2 loss between approaches, where one can see that the reconstructed HO_2 field shows a relatively small percentual decrease versus the situation without HO_2 uptake on cloud.

In the final version of our manuscript we propose to improve our methodology to account for SGS effects in the model by introducing an empirical SGS scaling factor to the HO_2 loss rate on cloud, as function of CC. This scaling factor can easily be tuned to match the simulated HO_2 concentrations towards the reconstructed field, representative for NM, hence providing an empirical parameterization to account for SGS effects.

An additional question posed by J.-F. Muller concerned our effective liquid and ice cloud radii in C-IFS, which were computed as function of the LWP and IWP, respectively. Additional to the description in the paper we should have mentioned that we limit $r_{e,cloud}$ to between 4 and 10 μm to avoid erroneous values. For ice radii, we do not limit the particle radius. Examining the distribution in C-IFS reveals we obtain values up to 45 μm maximum. Figure 3 of this response shows a zonal mean r_e for both cloud and ice to allay fears of very large values in our study.
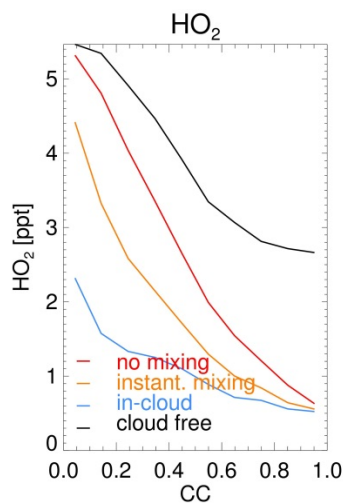

Figure 1. The average HO_2 volume mixing ratios at 800hPa for 1 April 2008 in C-IFS as sampled from instantaneous 6-hourly fields, and binned for cloud fraction ranges of 0.1. Colour key: Black: No HO_2 uptake in cloud is assumed ($HO_{2,free}$), orange: HO_2 uptake reaction scales with cloud fraction ('instantaneous mixing'), blue: HO_2 uptake reaction is not scaled, i.e. modeled HO_2 is representative for the value within the cloudy fraction ($HO_{2,cloudy}$) and red: Reconstructed HO_2 according to eq. 1, as best estimate for a 'no-mixing' assumption.
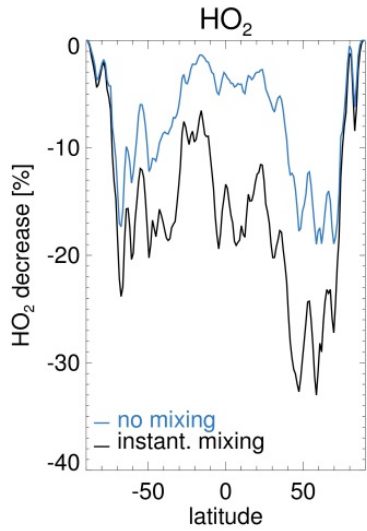
Figure 2. The zonal mean percentual decrease of HO_2 at 800hPa for 1 April 2008, on the assumption of instantaneous mixing (black) and no mixing (blue), as compared to the reference situation without any cloud HO_2 uptake.
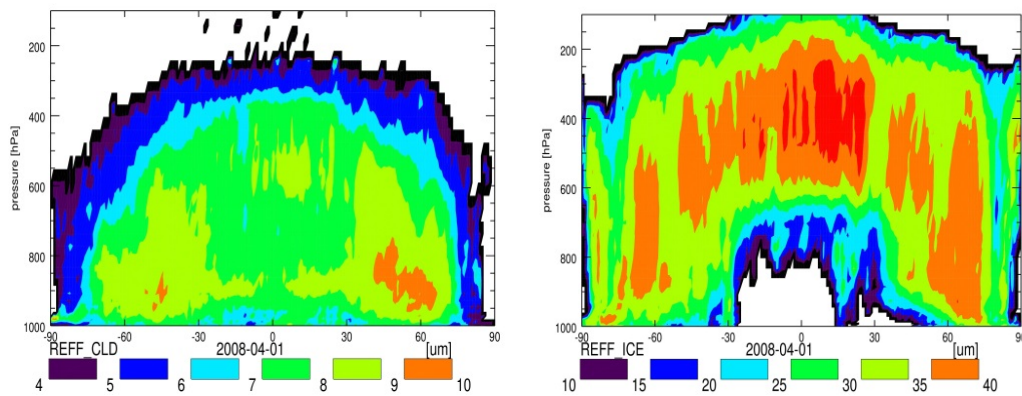

Figure 3. The zonal mean effective radius of liquid (left) and ice (right) cloud particles on April 1st 2008, given in μm.