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Transport and chemical transformations influenced by shallow cumulus over land

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Abstract. The distribution and evolution of reactive species in a boundary layer characterized by the presence of shallow cumulus over land is studied by means of two large-eddy simulation models: the NCAR and WUR codes. The study focuses on two physical processes that can influence the chemistry: the enhancement of the vertical transport by the buoyant convection associated with cloud formation and the perturbation of the photolysis rates below, in and above the clouds. It is shown that the dilution of the reactant mixing ratio caused by the deepening of the atmospheric boundary layer is an important process and that it can decrease reactant mixing ratios by 10 to 50 percent compared to very similar conditions but with no cloud formation. Additionally, clouds transport chemical species to higher elevations in the boundary layer compared to the case with no clouds which influences the reactant mixing ratios of the nocturnal residual layers following the collapse of the daytime boundary layer. Estimates of the rate of reactant transport based on the calculation of the integrated flux divergence range from to -0.2 ppb hr^{-1} to -1 ppb hr^{-1}, indicating a net loss of sub-cloud layer air transported into the cloud layer. A comparison of this flux to a parameterized mass flux shows good agreement in mid-cloud, but at cloud base the parameterization underestimates the mass flux. Scattering of radiation by cloud drops perturbs photolysis rates. It is found that these perturbed photolysis rates substantially (10–40%) affect mixing ratios locally (spatially and temporally), but have little effect on mixing ratios averaged over space and time. We find that the ultraviolet radiance perturbation becomes more important for chemical transformations that react with a similar order time scale as the turbulent transport in clouds. Finally, the detailed intercomparison of the LES results shows very good agreement between the two codes when considering the evolution of the reactant mean, flux and (co-)variance vertical profiles.

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