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Clouds, photolysis and regional tropospheric ozone budgets

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Abstract. We use a three-dimensional chemical transport model to examine the shortwave radiative effects of clouds on the tropospheric ozone budget. In addition to looking at changes in global concentrations as previous studies have done, we examine changes in ozone chemical production and loss caused by clouds and how these vary in different parts of the troposphere. On a global scale, we find that clouds have a modest effect on ozone chemistry, but on a regional scale their role is much more significant, with the size of the response dependent on the region. The largest averaged changes in chemical budgets (± 10 – 14%) are found in the marine troposphere, where cloud optical depths are high. We demonstrate that cloud effects are small on average in the middle troposphere because this is a transition region between reduction and enhancement in photolysis rates. We show that increases in boundary layer ozone due to clouds are driven by large-scale changes in downward ozone transport from higher in the troposphere rather than by decreases in in-situ ozone chemical loss rates. Increases in upper tropospheric ozone are caused by higher production rates due to backscattering of radiation and consequent increases in photolysis rates, mainly $J(\text{NO}_2)$. The global radiative effect of clouds on isoprene, through decreases of OH in the lower troposphere, is stronger than on ozone. Tropospheric isoprene lifetime increases by 7% when taking clouds into account. We compare the importance of clouds in contributing to uncertainties in the global ozone budget with the role of other radiatively-important factors. The budget is most sensitive to the overhead ozone column, while surface albedo and clouds have smaller effects. However, uncertainty in representing the spatial distribution of clouds may lead to a large sensitivity of the ozone budget components on regional scales.

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