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Development of a reduced aqueous phase chemistry mechanism

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Recent model studies have implicated the necessity of more complex aqueous phase processes to be considered in future higher scale chemistry transport models (CTMs). Important chemical cloud effects are mainly not yet considered or less represented in currently available regional scale CTMs. To this end, a mechanism reduction of the detailed aqueous phase chemistry mechanism CAPRAM 3.0i (Chemical Aqueous Phase RAdical Mechanism, Herrmann et al. [2005]) with about 777 reactions have been performed to develop simplified mechanism with less than 250 processes. For the mechanism reduction manual methods including detailed process investigations and automatic techniques [see Mauersberger, 2005] were applied. Both investigations have been done in order to provide a less computationally intensive mechanism which is operational in higher scale CTMs and accurately represents the main chemical aqueous phase processes.

The results of the manual reduction have been compared with the output of an automatic reduction. This comparison showed a quite good agreement. Based on the restrictions of both reduction methods, a final reduced mechanism was derived which describes the main characteristics of inorganic and organic aqueous phase processes occurring in tropospheric warm clouds. With less than 200 reactions, the reduced mechanism is nearly a factor of 4 smaller than the detailed CAPRAM 3.0i mechanism. Most of the chemical reduction potential has been realised in the organic chemistry with 393 unimportant reactions. Moreover, the number of aqueous phase species decreased from 380 in the full CAPRAM 3.0i mechanism to 130 in the final reduced version. Furthermore, 11 unimportant phase transfer processes and 36 insignificant chemical equilibriums have been identified according to their minor relevance for the preselected reduction key species. The calculated percentage deviations between the full and reduced mechanism are mostly below 5% for the most important target compounds.

Additionally, numerical sensitivity tests have been performed focusing on the relevance of both the relative and absolute integration error tolerances for an accurate and efficient modelling. The sensitivity studies have shown that cloud formation and particularly evaporation periods are circa three times more computationally intensive than in cloud conditions. This indicates the requirement for sufficiently accurate tolerance levels particularly there. Comparisons of the required CPU times between the full and final reduced mechanism showed reductions of approximately 40%.

Prospectively, the final reduced aqueous phase mechanism represents the basis for studying chemical cloud effects on regional scale with future CTMs and will be important for a better understanding of the multiphase aerosol cloud processing effects on regional scale as well as the interpretation of field data.