Modeling of approaches for describing non-ideal solutions and their application in process studies of atmospheric multiphase particles: Comparison with LACIS experimental data

Ahmad J. Rusumdar,R.Wolke, J.Voigtländer, F.Stratmann

Leibniz Institute for Tropospheric Research, Leipzig , Germany (rusumdar@tropos.de)

Key words: Aerosol-cloud interactions, Thermodynamic modeling, activity coefficients, surface tension, hygroscopic growth, cloud droplet activation

A cloud is a multiphase system comprising a gaseous phase, a particulate phase, and an aqueous phase. To assess the role of cloud chemistry and interactions between gases, cloud droplets and aerosol particles have to be taken into account in numerical cloud chemistry models. Decades of cloud microphysical research have not provided conclusive understanding of the physical processes responsible for droplet growth. The physical state and structure of tropospheric aerosol particles is still largely unknown despite their importance for cloud formation, for multiphase and heterogeneous chemistry in and on aerosol particles. Growth and chemical changes in multi-component aerosols are under investigation for a longer period of time, but the results are still heterogeneous. Modeling of such multiphase processes, the partitioning of species between vapor and aqueous phases, are considered as equilibrium processes describable by equilibrium constant corrected for temperature dependence and non-ideality in deliquescent particles. More frequently experiments as well as modeling studies are paying attention on either hygroscopic growth (RH < 100%) or cloud droplet formation (RH > 100%) regions. Variables of the Köhler equation are important for the low and high RH regions. Indeed, activity coefficients are more significant to predict hygroscopic growth (RH < 100%), since the droplets are far from ideal solution limit. To throw light on these issues, aiming with the improved description of such complex systems, different activity coefficient calculation methods has been implemented into the Spectral Aerosol Cloud chemistry Interaction Model (**SPACCIM**) [1], in addition, to compare with LACIS (Leipzig Aerosol Cloud Interaction Simulator) [2], experimental data.

The parcel model SPACCIM, has been developed for the description of cloud microphysical processes with the consideration of complex multiphase chemistry for

a size-resolved particle/drop spectrum. Microphysics is coupled with a chemical model, so meteorological and microphysical parameters needed for the chemistry can be taken from the microphysical model. The simulations were carried out for a meteorological scenario in which an air parcel moves along a pre defined trajectory including three cloud passages and intermediate aerosol state at a 90 % relative humidity level for three cases: (1) without feedback between the microphysical and chemical model and without consideration of non-ideal solutions; (2) without feedback, but with consideration of non-ideal solutions; (3) with feedback and consideration of ideal solutions.

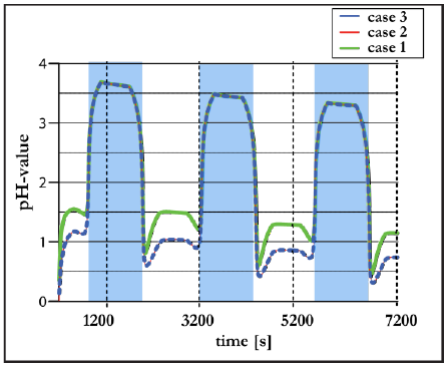

Figure 1. pH value- during the three cases

The non-ideality of highly concentrated solutions is described by activity coefficients. For the computation of activity coefficients the coupled models LIFAC [3], Ming & Russell [4] were implemented. The new SPACCIM version is able to simulate the physico-chemical processes of the particles/droplets also at lower relative humidities. The second model FPM/FLUENT is applied to model experiments which were performed on LACIS (Leipzig Aerosol Cloud Interaction Simulator) [2]. The output profiles of the FLUENT/FPM models, has been taken as input, for the meteorological scenario. While prescribing the microphysical conditions in to the model, the simulations were preformed. For comparison all the thermo dynamical properties, activity models, surface tension parameterization were synchronized.

References

[1]. Wolke, R., et al, *Atmospheric Environment*, 39 4375-4388, (2005).

[2].Stratmann.F et al., *J.Atmos.Ocean.Tech*, 21,876-887, (2004)

[3]. Yan, W. et al, *Fluid Phase Equilibria*., 162, 97-113, (1999)

[4].Ming, Y., Russell, L.M., *AIChE J*., 48, 1331–1348, (2004)