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Application of several activity coefficient models to water-organic-electrolyte aerosols of atmospheric interest

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Abstract. In this work, existing and modified activity coefficient models are examined in order to assess their capabilities to describe the properties of aqueous solution droplets relevant in the atmosphere. Five different water-organic-electrolyte activity coefficient models were first selected from the literature. Only one of these models included organics and electrolytes which are common in atmospheric aerosol particles. In the other models, organic species were solvents such as alcohols, and important atmospheric ions like NH_4^+ could be missing. The predictions of these models were compared to experimental activity and solubility data in aqueous single electrolyte solutions with 31 different electrolytes.
Based on the deviations from experimental data and on the capabilities of the models, four predictive models were selected for fitting of new parameters for binary and ternary solutions of common atmospheric electrolytes and organics. New electrolytes (H^+, NH_4^+, Na^+, Cl^-, NO_3^- and SO_4^{2-}) and organics (dicarboxylic and some hydroxy acids) were added and some modifications were made to the models if it was found useful. All new and most of the existing parameters were fitted to experimental single electrolyte data as well as data for aqueous organics and aqueous organic-electrolyte solutions. Unfortunately, there are very few data available for organic activities in binary solutions and for organic and electrolyte activities in aqueous organic-electrolyte solutions. This reduces model capabilities in predicting solubilities.

After the parameters were fitted, deviations from measurement data were calculated for all fitted models, and for different data types. These deviations and the calculated property values were compared with those from other non-electrolyte and organic-electrolyte models found in the literature. Finally, hygroscopic growth factors were calculated for four 100 nm organic-electrolyte particles and these predictions were compared to experimental data and to predictions from other models.

All of the newly fitted models show good agreement with experimental water activity data in binary and ternary solutions. One of the models is for activities of non-electrolytes only, but the other three models show quite small deviations from measured electrolyte activities. Because there were not enough experimental data for organic and electrolyte activities, some models show bigger deviation for mutual deliquescence relative humidities of organic-electrolyte particles, but calculated growth factors for liquid droplets are quite close to the experimental data. Even in cases with

somewhat bigger deviations, the results can be considered satisfactory, because they were calculated based mainly on the predictive properties of the models.

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