Review of the paper
"Determination of interfacial parameters of a soluble particle
in a nonideal solution from measured deliquescence and efflorescence humidities",
by O. Hellmuth and A. K. Shchekin,
Atmos. Chem. Phys. Discuss., 14, 22715–22762, 2014
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Recommendation: accept with minor editorial revisions

The paper by O. Hellmuth and A. K. Shchekin is devoted to the theory of aerosol deliquescence and efflorescence. The subject of this study is within scope of the topics considered in the journal ACPD-ACP and is very important for the models of various scales, including climate models, and for the assessments of global climate change. Deliquescence of aerosol determines its optical thickness, and thereby, the direct aerosol affect. It also influences activation of CCN into cloud drops and determines the 1st and 2nd indirect aerosol effects via cloud drops and, after subsequent freezing – several other indirect aerosol effects (e.g., Lohmann and Feichter, 2005).

The activated drops participate in the cloud dynamical circulation. When they are brought by the downdrafts near the cloud edges below cloud bottom, they may penetrate into the very dry environment. This may cause the reverse effect: evaporation of the drops to the sizes of CCN with strongly supersaturated solution concentration, which may cause efflorescence and transformation of the aerosol particles to the state close to the original dry or slightly wetted CCN. When such particles are brought again into the cloud by the updrafts, they grow again, may be activated and influence cloud microphysical and optical properties. With account for significant variability of aerosol over the globe, it is clear that any reliable estimates of aerosol on clouds and climate change cannot be done without the good and precise theories of aerosol deliquescence and efflorescence.

Such a theory is developed in the paper by Hellmuth and Shchekin. Based on the general principles of thermodynamics and kinetics and using previously developed by the authors generalizations of the Gibbs–Kelvin–Köhler (GKK) equation of the theory of nucleation and of the Ostwald–Freundlich (OF) equation of the theory of solutions, the authors apply these theories to the study of deliquescence and efflorescence. This allows authors to calculate several fundamental characteristics of these processes, and the whole cycle of their temporal evolution between the dry and liquid states under various conditions. Several new generalizations of the theory were performed when developing this theory, in particular, account for non-ideality of solutions, and account for the size effects of aerosol particles, which allows to perform calculations for very small particles down to nano-sizes. When developing this theory and its applications, the authors demonstrate the outstanding, really fluent and brilliant, skills in mathematical tools and very deep understanding of the physical processes that govern deliquescence and efflorescence.

Manuscript Evaluation Criteria. From the point of view of **Scientific Significance, Scientific Quality, and Presentation Quality**, the present manuscript can be estimated as very close to the category **Excellent (1).** Answering the 15 questions of the manuscript evaluation, I would give the following answers: 1) the paper addresses relevant scientific questions within the scope of ACP; 2) the paper present novel concepts, ideas, tools, and data; 3) the substantial conclusions are reached; 4) the scientific methods and assumptions are valid and clearly outlined; 5) the results are sufficient to support the interpretations and conclusions; 6) the description of experiments and calculations is sufficiently complete and precise to allow their reproduction by fellow scientists; 7) the authors give proper credit to related work and clearly indicate their own new/original contribution; 8) the title clearly reflect the contents of the paper; 9) the abstract provide a concise and complete summary; 10) the overall presentation well structured and clear; 11) the language is fluent and precise; 12) mathematical formulae, symbols, abbreviations, and units are correctly defined and used; 13) no parts of the paper (text, formulae, figures, tables) should be clarified, reduced, combined, or eliminated; 14) the number and quality of references are appropriate, although a few older and newer references can be added (see specific remarks); 15) the amount and quality of supplementary material is

appropriate; to make the paper more readable and clear, the authors moved most of the previous mathematical tools to supplement; and an interested reader can easily retrieve this supplement.

Specific remarks (these should not be considered as mandatory, up to the authors' discretion)

1) It would be worthy to add a few introductory sentences (similar to those at the beginning of this review) showing importance of deliquescence and efflorescence for atmospheric models and processes and especially for climate models. This would emphasize the importance of this paper devoted to studies of these processes and for assessment of climate change.

2) Classical nucleation theory usually uses a capillary approximation. Numerous attempts were made to improve or modify this approximation, especially for very small particles. The authors of this work introduce and use an alternative approximation of "disjoining pressure". However, its description is too short and not very clear. It would be worthy to add a few sentences on page 12 (22726) with explanation of the physical meaning of "disjoining pressure", why it occurs, how it acts and how it is related to the traditional capillary approximation. Is capillary approximation a particular case of the "disjoining pressure" or there is no relations between them?

3) Similar remark. On page 19 (22733), eqs. 7 and 8, the 2 "cost functions" are introduced. Their introducing is also somewhat formal, and some comments are desirable here; it is "cost of what?", and what do they characterize?

4) As I could understand, the baseline calculations in this work were performed at T = 298 K. Laboratory measurements show that there is a substantial temperature dependence of the deliquescence and efflorescence (e.g., Seinfeld and Pandis, 1998; Cziczo and Abbatt, 1999; Oatis et al., 1998; Xu et al., 1998), which is caused, in particular, by the temperature dependence of the surface tensions. In a recent work by Khvorostyanov and Curry, (2014, Chapter 11 therein), it was shown that these lab data can be reproduced based on extended CNT, and using appropriate reference points $\sigma(T_0)$ and temperature gradients of the surface tensions, $(d\sigma/dT)$. I guess, the theory of Hellmuth and Shchekin can be extended into a wider temperature range, and a short comment in Conclusions would be helpful on the possible T-extensions of this theory and appropriate necessary choice of the gradients $(d\sigma/dT)$.

5) The authors concentrated in this work mostly on NaCl. Another aerosol, ammonium sulfate, may play an important role in formation of the upper-level clouds like cirrus. If the authors plan to perform s similar study for ammonium sulfate (as Gao et al, 2006, 2007, made in 2 separate papers), it might be worthy to add such comment in Conclusions.

6) Can any recommendation be given, based on this work, for application of this theory in cloud or climate models? If so, this could be added in Conclusions.

7) p. 23, 1st line. Is written "mass fanction", misprint. Should be "mass function".

The remarks above are mostly editorial and do not diminish the value of this paper, which is certainly one of the strongest works ever done in this area, and the paper unconditionally deserves to be published in the journal Atmospheric Physics and Chemistry.

References

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