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Interactive Comment

Interactive comment on "Application of osmolality for the determination of water activity and the modelling of cloud formation" *by* G. Kiss and H.-C. Hansson

Anonymous Referee #2

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This paper presents a novel method for accurately calculating critical supersaturations and critical droplet diameters. The use of osmolality derived water activities eliminates the need for standard estimations and assumptions in Kohler theory. I recommend the publication of this paper in ACP, however, numerous questions should be addressed and issues clarified.

1. The paper mentions numerous times the effect of assuming that the van't Hoff factor for dicarboxylic acids is 2 or 3 leads to large errors. On page 7671, line 23, the authors state, "It is often assumed, for example, that i=2 or i=3 for dicarboxylic acids..."



This statement is not supported with references, however, and may not be the case. With the possible exception of a few cases, it has been widely assumed that i=1 for dicarboxylic acids. As the authors point out, this is easily demonstrated with a back of the envelope calculation with literature pKa values for the acids in question.

2. For readers not familiar with osmolality, equation 4 appears with no description of how it was derived or whether it came from the Kiyosawa paper. Also, does this approximation (Eq. 4) break down under any circumstances?

3. A main point of the paper is the applicability of using osmolality derived water activities in real-world situations or real atmospheric samples. Yet all the examples given are for single component particles. Furthermore, there is no mention of how to apply this to real world problems. I assume that osmolality is a bulk measurement and could only be used on large samples such as fog water and not on individual particles or droplets. Air masses, depending on their history can have many different types of particles and it seems that this technique is only useful for ensemble averages, rather than individual particles. This would seem to strip much of the predictive power from this method. For example, there are numerous techniques for measuring compositions of individual aerosol particles, however, if we do not know, or can't measure the osmolality of these particles (or droplets), then this new technique, while accurate, does not help us predict whether this particle will be an efficient CCN. I hope that the authors could comment on this.

4. The largest corrections to the calculated critical supersaturations using osmolality occur for the smallest and most electrolytic solutions, however, osmolality does not take into account the surface tension of the droplet. The corrections for sucrose are fairly small and the corrections would presumably also be fairly small for organic rich particles. Does the somewhat unknown and potentially important surface tension effect reduce the usefulness of this method in dealing with organic rich particles?

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Many dependent clauses were not separated by commas. For example: Page 7668, line 26 "However, the magnitude..." Page 7669, line 12 "In this equation, aw accounts..." Page 7670, line 09 "has been shown that, in addition to inorganic salts, organic..." etc.

Page 7670, line 2-3 "with cloud condensation nuclei counters."

The quality of the figures could be improved.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 7667, 2004.

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