Response to J. Rissler

We thank J. Rissler for the review, constructive comments and suggestions for improvement of our manuscript. Detailed responses to the individual comments (including additional information and figures from the revised manuscript) are given below.

(Referee comments in *italics*, the responses in plain font)

Comments and suggestions:

The main concern regarding the work is weather the data was inverted before using the data – taking into account the effect of the DMA transfer function, etc. From the text it is not clear weather this is considered or not. There are some indications that the data was inverted before applying the approach here described, but it is not obvious weather this was done both for the H-TDMA data and the CCN counter data. If this was considered, the authors need to write this more specifically, describe how this was done and point out the importance of this. If data was not inverted, it would improve the work considerably to do this. At least the authors need to comment on how this would affect the results and make some quantification of the effect.

Responses and Revisions

The data used in this paper have been inverted as follows:

Page 6 line 159-162

"The HTDMA calibration and data inversion accounted for the counting efficiency of the condensation particle counter (CPC) as well as for the transfer function of the DMA. Further details about the measurement campaign, techniques and conditions are given by Massling et al. (2009)."

Page 14 line 402-406

"The size-resolved CCN measurements were performed following method II ("Dd scan" at constant S), and the data were recorded and processed as detailed by Rose et al. (2010a), except that the DMA transfer function correction was not applied, because it is based on the assumption of perfect internal mixing (Sect. 2.3.1.3 in Rose et al., 2008)."

Comments and suggestions:

Introduction: In the introduction previous attempts to parameterize the rate of external mixture from CCN counter measurements are discussed. Since this work focus on H-TDMA data, as much as on CCN counter data, I think a discussion about previous studies that have made attempts to describe the rate of external mixture from the particle hygroscopic growth measured by the H-TDMA would be in place. For example, in the inversion algorithm presented in the work by Stolzenburg and

McMurry (1988) multiple hygroscopic modes could be fitted and a parameter describing the broadening of each mode given. This is also the case in the recent work by Gysel et al. (2009). I suggest that these studies should be mentioned and discuss the advantages with the concept here introduced. Furthermore, in several previous studies presenting H-TDMA data attempts to describe the rate of external mixture have been made by making bi- or tri-modal fits of the wet distribution, describing the growth and particle fraction of each mode separately (references in Swietlicki et al., 2008).

Responses and Revisions

We included the work of Stolzenburg and McMurry (1988) , Swietlicki et al. (2008) and Kammermann et al. (2010) in the introduction:

Page 2 line 47-51:

"Early and recent HTDMA studies have already presented distributions of diameter growth factors, which are also related to particle hygroscopicity, and addressed the relation to aerosol mixing state (Stolzenburg and McMurry 1988; Swietlicki et al., 2008; Kammermann et al., 2010)."

Comments and suggestions:

Page 1008, row 8: The notations N(kappa) and N(kappa) are confusingly similar and since the absolute number concentration is not used I suggest not to introduce this quantity N(kappa).*

Responses and Revisions

In the revised manuscript, the absolute value and symbol were excluded.

Comments and suggestions:

Page 1008, row 19: Van not Hoff factor ! Van't Hoff factor

Responses and Revisions

Corrected

Comments and suggestions:

Page 1009, row 7-9: The authors argue that the kappa-distributions can be described by log normal distributions based on the fact that particle size distributions are lognormal. For me it is not clear why this is. However, later it is shown that this is a good hypothesis. In Stolzenberg and McMurry (1988) normal distributions were used *to describe the shape of each hygroscopic mode.*

Responses and Revisions

We added quantitative measures for the fitted distributions:

Page 6 line 169-170:

"… indicates a lognormal mode in the hygroscopicity distribution (lognormal fits yield R2 of 0.99) …"

For the distribution function, we also clarify it (page 14 line 434-438) : "other fit functions such as normal or Gamma distributions, power laws, etc. (Deirmendjian, 1969; Pruppacher and Klett, 1997; Seinfeld and Pandis, 2006) or multiple lognormal modes may be equally or better suited depending on aerosol composition and the range and resolution of measurement data, which should be explored in futures studies."

Comments and suggestions:

Page 1011, row 11-.. : If easily done it would be really interesting to se a comparison of the kappa-functions derived from H-TMDA and CCN counter measurements made in parallel for aerosol with some external mixture. This would show how useful the concept is when comparing the particle's water uptake at sub and super saturations, possibly giving information on limited solubility, varying osmotic potential etc.

Responses and Revisions

This is a good suggestion and we would like to pursue on such studies.

Comments and suggestions:

Page 1011, eq. 7: In Petter and Kreidenweiss (2007) this approximated form of the Köhler equation is recommended to be used for kappa >0.2 only. For lower 'kappa' an error is introduced using the approximation, especially for small particles activating at high supersaturations. The approximate form of the Köhler equation is in many studies used also for kappa <0.2, but a comment about this would be in place, and a short note about which effect this have on the derived kappa-distribution.

Responses and Revisions

Yes, these equations are only approximation. We clarify this and explain how to numerically solve the Koehler equation for an accurate solution.

Page 8 line 213-217:

"the critical value of the third parameter can be determined by numerical calculation of the maximum point in the corresponding Koehler model curve of CCN activation (Rose et al., 2008). … Alternatively, the critical parameter values κ_c, $D_{d,c}$ and S_c can be approximated as follows (Petters et al., 2007, Rose et al., 2008)…"

For the bias of the approximation, we decide to address it thoroughly in another coming paper.

Comments and suggestions:

Page 1011, section 2.3: In this section it is not explained how monodisperse particles are selected before fed into the CCN counter. Most often this is made with a DMA, but it is not described in this section.

Responses and Revisions

Yes, a DMA is used. We revised the manuscript as follows:

Page 7 line 197-199:

"In such measurements, monodisperse dry aerosol particles with a diameter D_d are usually selected by a DMA, and the monodisperse aerosol is fed into a CCNC and into a CPC operated in parallel to measure the number concentrations of CCN".

Comments and suggestions:

Page 1013, row 5: "Method I is easier to interpret: : :" Seems like a subjective comment.

Responses and Revisions

We put more discussions to explain the differences of the two methods which makes it sound more objective:

Page 9 line 237 - 271 and a new Fig.6:

"In principle, the two methods are equivalent with regard to probing the surface … because many of the recently reported size-resolved CCN field measurements used this approach (varying D_d at a constant S)."

Comments and suggestions:

Page 1013, row 25: Here I guess that the reference here should be Rissler et al (2005) - if refereeing to CCN predictions.

Responses and Revisions

It is cited as an exemplary hygroscopicity study with good quality control.

Comments and suggestions:

Page 1014, row 6-12: Svenningsson et al. (2008) made an attempt to separate the effect of the DMA transfer function with the effect of low solubility and external mixture in CCN counter data. I suggest referring to this work.

Responses and Revisions

We added this reference in the revised manuscript.

Comments and suggestions:

Page 1014, row 19-22: Specify what is meant by "ideal measurement conditions". Fully internally mixed particles/instant dissolution/only singly charge particles or "ideal" DMA's?

Responses and Revisions

The ideal conditions means perfect particle generation, measurement and data analysis, e.g. all particles are the same, with the same chemical composition, same shape and mixing state; no multiply charged particles; the DMA is "ideal" with a delta transfer function and the particles selected by such DMA can still be accurately detected by the following CPCs. Or the data inversion and correction methods are good enough to count and correct all these effects.

Comments and suggestions:

Page 1016, row 21: hygrocopicity ! hygroscopicity

Responses and Revisions

Corrected.

Comments and suggestions:

Page 1017, row 1-.. : This size dependence is most often observed in atmospheric studies.

Responses and Revisions

This is better than our last statement, but we found that actually such distribution is not always true, especially for clean areas like Amazonia rainforest, so we corrected it as,

Page 12 line 347-348:

"Similar CCN efficiency spectra have been recorded in polluted megacity air (Sect. 4.2, Rose et al., 2010a)."

Comments and suggestions:

Page 1020, row 28: Wrong order of references.

Responses and Revisions

Corrected