

Dear Prof. Covert,

We greatly appreciate the referees for their valuable comments and suggestions. The manuscript has been revised accordingly. Point-by-point responses to all the comments along with the revised manuscript and figures have been uploaded.

Best Regards
Chunsheng Zhao

Response to Anonymous Referee #1:

10. Is the overall presentation well structured and clear? From line 22 Page 4091 to line 5 page 40929(13 lines), more models were introduced, in which there were six lines to introduce the ADDEM model, but it was not used in this work. In fact, model ISORROPIA II was used. It is better to give some words to describe why this model was chosen.

Response: Thanks for your comment. We have rephrased this paragraph. The introduction about the ADDEM model has been shortened. More sentences to describe the ISORROPIA II model have been added. The reason for choosing ISORROPIA II model in this work has been expounded in Section 3.3 in the text. These sentences have been revised as:

“Most models only consider how inorganic solutes influence the saturation vapour pressure as solute (Raoult effect). Topping et al. (2005a, 2005b) developed a model called ADDEM, which considers both the Raoult effect and the curvature effect of the organic/inorganic aerosol components. Chang et al. (2001) calculated the purely theoretical water content of ambient aerosols with the ISORROPIA model to adjust the discrepancy of PM₁₀ concentrations between the results measured by the β -gauge monitor and sampled by the hi-vol sampler. Hennigan et al. (2008) predicted the liquid water concentration with the ISORROPIA II thermodynamic equilibrium model, using mean sulfate, nitrate, and ammonium concentrations from the August 1999 Atlanta Supersite data. Though the model does not consider the effect of organics on water uptake, they thought that the high inorganic concentrations of PM_{2.5} suggest that the model results provide a reasonable estimate.”

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? There were too many abbreviations to be defined and used. Some abbreviations need not be defined, since they were not used frequently

Response: Thanks for the comment. The following rarely used abbreviations are removed from the text: HDMPS, DAASS, VGF, SMPS, PRD, AWS, CPCs.

Response to Anonymous Referee #2:

General comments:

Aerosol liquid water content (ALWC) is one of the key quantities, which are closely related to aerosol optical properties and climate effect and to chemical reactions that take place in aerosol particles. Although ALWC can be calculated using some thermodynamic equilibrium models, all models are highly simplified and have some limitations. A few methods have been reported to calculate ALWC from measurements. However, most of the previous methods are limited to RH lower than 90%. This paper proposed a method of calculating ALWC under RH 20-99% conditions. Size-resolved hygroscopic growth factors are obtained based on High Humidity Tandem Differential Mobility Analyzer (HH-TDMA) measurements and using the kappa-Köhler theory. ALWC values for the HaChi campaign are then calculated from the hygroscopic growth factors and particle number size distribution (PNSD). The ALWC values are compared with those from the ISOPROPIA II model. Impacts of RH and PNSD on the calculated ALWC values are studied.

I think this paper addresses relevant scientific questions within the scope of ACP. The methods used in this paper are novel. The results presented are definitely of interest and valuable. This paper is well structured and concise. The authors give proper credit to related work. I only have a few minor points and recommend publication of this paper in ACP after minor revisions.

Response: Thanks for the referee's comments.

Specific comments:

1. P4093, L4-5, "the RH measured"?

Response: We have changed “the RH measured” to “the RH values recorded”.

2. P4097, L20-22, why T used to calculate κ is set to 293K? Was this the average T ? Aerosol water is not pure water. Do we expect a surface tension coefficient much different from 0.0728 N m^{-1} ?

Response: Thanks for your comment. The room temperature of our container was set to 293K at the site during the campaign. An explanation has been added in the text. Some constituents of the atmospheric aerosol are surface active and can suppress surface tension. Use of a lower value of surface tension (the surface tension coefficient between water and air) would result in a decrease in the derived value of κ . However, this lower κ must then be applied in Eq. (2) along with the lower surface tension to calculate the size-resolved growth factor. Either approach, as long as κ and $\sigma_{s/a}$ are applied self-consistently, should yield estimates of growth factor that reproduce the measured data (Petters and Kreidenweis, 2007).

Citation: Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7(8), 1961-1971, 2007.

3. P4098, L10-11, the size-resolved growth factor is not explicit in Eq. (2). I think more explanations on the resolving process are needed.

Response: Thanks for your suggestion. We have added more information on the fitting and calculating process. This paragraph has been revised as :

“Liu et al. (2011) found that, for a certain particle diameter, the according growth factors varies with RH. The κ value, however, does not vary with RH. The HH-TDMA was used to measure the hygroscopic growth factors and the hygroscopicity parameter κ at four dry aerosol diameters (50nm/100nm/200nm/250nm) in this campaign. Calculations need to be made to derive a size-resolved hygroscopic growth factors. First, a 4 mode lognormal distribution is applied to fit the measured PNSD, yielding fitting parameters for each mode (Tareq et al., 2005). Afterwards, assuming that κ is constant within the same mode, the measured κ can be used to deduce the corresponding κ for each of the four modes of the reconstructed PNSDs. Considering the primary chemical component of the coarse particles is nearly hydrophobic, the κ for the coarse mode is assumed to be 0. Then the size-resolved κ can be calculated by combining the 4-mode fitting results with the measured κ values at certain particle diameters (Fig. 2). Finally, the size-resolved κ is substituted into Eq. (2) to calculate the size-resolved growth factors at 20~99% RH, respectively. The detail of the fitting and calculating process is described by Chen et al. (2012).”

4. Section 4.3.1 and Fig. 5, a fitted function connecting the relative ALWC and RH would be interesting and valuable.

Response: Thanks for the suggestion. A fitted function has been added in the text following your suggestion. The text has been rephrased as:

“To describe the relationship between relative ALWC and RH, a two-parameter fitted function is used with the relative ALWC and the ambient RH (Fig. 5):

$$\text{Relative ALWC} = a((1 - RH)^{bRH} - 1), \quad (7)$$

where the fitted parameters a and b are 0.91 and -0.78, respectively. The coefficient of determination R^2 is 0.99 approximately.”

The replotted Fig. 5 can be found below.

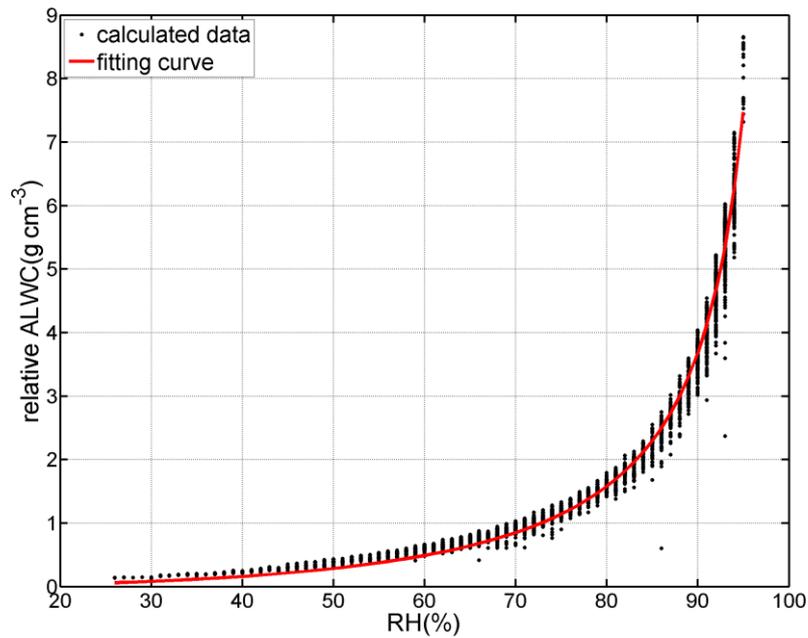


Figure 5. The relationship between relative ALWC and RH. Relative ALWC is defined as the mass of water absorbed by unit volume of dry aerosols. The black dots represent the relative ALWC calculated with ALWC and aerosol volume concentration. The red line shows the curve fitted with Eq. (7).

5. P4103, L2-3, the R^2 value of the Aitken mode under $RH > 90\%$ is 0.35, which is much larger than 0.2.

Response: Some more-hygroscopic particles in the Aitken mode absorbed much more water at RH higher than 90%. That's why the R^2 value of the Aitken mode under $RH > 90\%$ is larger than 0.2. The sentences have been added as:

“For $RH > 90\%$, the coefficient of determination of the Aitken mode particles is 0.32 and is much higher than the R^2 values under other RH conditions. The reason for the higher coefficient of determination is that the more-hygroscopic particles in the Aitken mode absorb much more water when RH is higher than 90%.”