

Reviewer: *This manuscript describes the hygroscopic properties of the aerosol during summertime in the North China plain. Hygroscopic growth for RH values between 90 and 98.5% are presented. Parameterizations of hygroscopic properties are carried out, and the diurnal variation of the aerosol is discussed and compared to modelled values. The data in the paper is of good quality (validated both with salt measurements and with intercomparison to LACIS) and the fact that the HH-TDMA provides data for $RH > 90\%$ is very nice, as this region is often left for speculation regarding solubility etc. in CCN closure studies which connect H-TDMA measurements at 90% with CCNC data. This makes the data especially relevant. I recommend this paper to be published after modifications and clarifications described below.*

Response: Thanks for the positive comments.

General comments

Reviewer: **) My main comment is that the paper is unnecessarily long. Certain parts could be excluded and in some cases a figure says more than a thousand words. It is better to focus on interpretation in the text, and only briefly describe the numbers in a figure or a table. If the description is too long, one tends to miss the main points. There are also a number of strange formulations that need to be rewritten. I recommend that you make an effort to shorten the manuscript, at least chapter 2, and I will give specific recommendations in the following detailed comments.*

Response: According to the comments from all of three referees, the chapter 2 “Data and methods” has been restructured and significantly shortened. Other chapters have also been trimmed. Following the detailed comments from referees, most contents of section 2.2.2 have been deleted. The chapters 2.2.3 have been shortened and restructured. The equations (1)-(4) and (6)-(10) have been removed.

Reviewer: **) You use the term “CCN activity” a couple of times in the text, but you never explain what you mean by it. Please do so.*

Response: Thanks for pointing out our vagueness in using the term “CCN activity”. In the revised paper, the term “CCN activity” has been replaced by more specific expressions given below.

Page 2994, line 3: the “cloud condensation nuclei (CCN) activity” has been replaced with the “ability of aerosol particles to activate and form cloud droplets”.

Page 2994, line 25: “The RH-dependency of κ could be crucial for the prediction of CCN activity”, has been replaced with “The RH-dependency of κ could be crucial for the prediction of CCN number concentration”.

Page 2996, line 15: the “CCN activity” has been replaced with “size resolved and bulk CCN activation properties (Deng et al., 2011)”.

Detailed comments:

Reviewer: *Page 2992, row 6: “dry diameter between 50 – 250 nm”, should be “dry diameters between 50 and 250 nm”.*

Response: This has been corrected in the revised paper.

Reviewer: Page 2992, row 18: Delete “, indicating a diurnal...”. It is essentially what you said in the first part of the sentence.

Response: This has been deleted as suggested.

Reviewer: Page 2992, row 22: “The high degree of...” This sentence is strange. You haven’t really defined “high”. I suggest simply writing that the fraction of NH particles was higher during night time.

Response: We have deleted the sentence “The high degree of...”, and simply mentioned that the fraction of NH particles was higher during night time.

Reviewer: Page 2993, row 27: “To understand...” I know what you are trying to say with this sentence, but it needs to be re-written, and preferably divided into two (or more) sentences. E.g. “the aerosol hygroscopic properties are very crucial since they describe the interactions of aerosol particles with water vapour” makes no sense, and what do you mean by “the CCN activity of aerosol particles and their number-size distribution”? Which size distribution are you referring to?

Response: In the revised paper, this sentence has been rephrased as “The aerosol hygroscopic properties are very crucial for the understanding of the aerosol effects on climate via their effects on clouds, since the hygroscopic growth measured at subsaturation is closely related to the ability of aerosol particles to activate and form cloud droplets (Swietlicki et al., 2008; Rissler et al., 2010).”

Reviewer: Page 2992, row 15: Use either “kappa” or “a single-parameter Köhler model”, but not both in the same sentence. I prefer the second alternative.

Response: In the revised paper, we use “a single-parameter Köhler model” in this sentence, as the reviewer suggested.

Reviewer: Page 2994, row 5: delete “(but less than...)”. It is true also for $RH > 100\%$.

Response: We have deleted “(but less than...)” in the revised paper.

Reviewer: Page 2994, row 6: “water uptake, hence” should be “water uptake, and hence”.

Response: This has been corrected.

Reviewer: Page 2994, row 17: There are several other descriptions of aerosol hygroscopicity and CCN modeling, recently overviewed in Rissler et al. (2010).

Response: A sentence “Moreover, Rissler et al. (2010) recently overviewed several models which describe the aerosol hygroscopicity and the CCN activation” has been added in the revised paper.

Reviewer: Page 2994, row 23: “by a factor of 4-6”. Explain that this factor is linearly proportional to the number of soluble entities per dry volume (scaled for water molar mass and density in the case of Petters’ kappa). This is helpful

information when interpreting changes in kappa. Because I presume that the “increased hygroscopicity parameter” is kappa or rho_{ion} (which is exactly the number of soluble entities per dry volume unit).

Response: In the revised paper, this explanation has been added as suggested.

Reviewer: Page 2994, row 25: “validations of the...”. If you count CCN closure studies as a validation, then there are a lot of reports. But if you are talking about only H-TDMA data, then you are correct. Please clarify.

Response: We have clarified in the revised paper that we only count the H-TDMA data.

Reviewer: Page 2995, row 10: “The heterogenouses..”. I do not understand this sentence. Firstly you say “The heterogenous mixtures”. Which mixtures are you talking about? Typical ambient aerosol particles? Then you say “hence, detailed hygroscopic properties provide important information on the aerosol mixing state”, but there is no causality here as far as I can see? Please rephrase this.

Response: We have rephrased these sentences as: “The hygroscopicity of aerosol particles can be considered as an agent of chemical composition. A Humidified Tandem Differential Mobility Analyser (H-TDMA) is one of the few instruments capable of providing information regarding the mixing state in terms of hygroscopicity of the atmospheric aerosols (Swietlicki et al., 2008). Different modes of hygroscopic growth of ambient aerosols are often observed in the H-TDMA measurements, indicating an external mixture in terms of hygroscopicity (Swietlicki et al., 2008).”

Reviewer: Page 2996, row 4: Delete “in this paper as well”.

Response: “in this paper as well” has been deleted.

Reviewer: Page 2997, row 27: “The direct humidification has its technical limits as well.” Please explain what you mean.

Response: We have added an explanation and revised this sentence as: “The direct humidification has its technical limit as well, because the water vapour transport through semi-permeable membranes is limited (Hennig et al., 2005).”

Reviewer: Chapter 2.2.2. This subchapter can be deleted altogether. It is enough with a reference to Petters and Kreidenweis, as it is all described there.

Response: The chapter 2.2.2 has been deleted in the revised paper.

Reviewer: Page 3002, row 18: You can delete the reference to Sjogren. Possibly a reference to the ZSR assumption would be in order, but this follows from basic physics.

Response: The reference to Sjogren et al. (2008) here has been deleted.

Reviewer: Page 3003, row 1: missing a superscript “3” after GF in the equation, it looks like?

Response: Yes, a superscript “3” was omitted in typesetting. We have added it in the revised paper.

Reviewer: *Chapter 2.2.3 in general could be significantly shorter. You can refer to Gysel et al. (2009) regarding the inversion and that would be enough. You do not need to write equations 6, 7, 9, 10, 11 and 12. It would be enough to state that you recalculated GF-PDFs to kappa_PDFs, that you use 3rd moment average values etc. I realize that you put effort into this, but my opinion is that it is a bit too much for this paper.*

Response: This chapter has been significantly shortened in the revised paper. We restructured this part following the detailed comments from *Referee 1* and *Referee 3*. The equation 6, 7, 9, 10, 11 and 12 have been eliminated in the revised paper.

Reviewer: *Page 3004, row 1: Why were the fluctuations lower for higher RHs? Also this subchapter could be described with a few sentences. Simply state your RH precision in the measurements, that GFs were recalculated using kappa and that the GF-PDFs were recalculated using the gamma-model (with reference).*

Response: By this sentence we intended to provide the deviations of the actual RHs from the set values. The using of the word “fluctuations” might be inappropriate here. We have rephrased this sentence as :”Over the whole summer campaign, the deviations of actual RH (measured by the DPM and calibrated using the ammonium sulphate particles) from the set value were smaller than 1.5% for 90% RH, while for 95% and 98.5% RH the deviations were normally within $\pm 0.5\%$ from set values.” This subchapter has been significantly shortened in the revised paper, following the referees’ comments.

Reviewer: *Page 3009, row 18 “via kappa-Köhler model” should be “via the kappa-Köhler model”.*

Response: This has been corrected.

Reviewer: *Page 3009, row 28: Delete “The aerosol mixing state will be discussed in the following sections”.*

Response: This sentence has been deleted.

Reviewer: *Page 3010, row 18: Delete “In other words, hygroscopicity...”. This is already stated a number of times before.*

Response: This sentence has been deleted.

Reviewer: *Page 3010: “Significant hygroscopic diameter growth behaviors were observed”. What does this mean?*

Response: This sentence has been deleted.

Reviewer: *Page 3013, row 9: Delete “we will present a...”*

Response: This has been deleted in the revised paper.

Reviewer: Page 3013, row 13: Delete this sentence.

Response: This sentence has been deleted in the revised paper.

Reviewer: Page 3014, row 7: Delete “i.e. the variance was low”. You already said this in the first part of the sentence.

Response: This has been deleted in the revised paper.

Reviewer: Page 3016, row 24: “It is worth to note that...” Is this because of higher contents of mineral dust?

Response: Yes, it is. We have added a sentence in the revised paper: “The decrease in hygroscopicity for large particles is because of higher contents of mineral dust.”

Reviewer: Page 3018. You use “thus” both on row 20 and 22. Looks a bit strange.

Response: The “thus” on row 20 has been replaced with “so that”. The sentence on row 22 has been rephrased and divided into two sentences as: “For case 4, to disable the entrainment of aerosol from aloft in the morning, the mixing height was fixed to 1000 m (see Fig. 9 (a)) base on case 3. The evolution of aerosol properties in the air parcel were therefore driven by the aging processes e.g., coagulation, condensation of secondary organic and inorganic species, and photochemical oxidation.”

Reviewer: Page 3020, row 12: “We should also notice that...”. I do not understand what you mean.

Response: This vague sentence has been rephrased as: “It is important to note that in the simulations we assume the aerosols above the mixing layer to be an internal mixture of sulphate, OC and BC. The reasons are that these lofted aerosol particles have undergone longer aging processes compared to those in the surface layer near emission sources and that the aging of lofted aerosol particles may be accelerated by cloud processes.”

Reviewer: Page 3020, row 16: What do you mean by “more direct”?

Response: The “direct” has been deleted in the revised paper. “...more three-dimension measurements of aerosol mixing state (e.g., Pratt and Prather, 2010) could be valuable for our scientific understanding”.

Reviewer: Page 3020, row 29: “formation”. Maybe this is OK, but I would prefer “condensation”, since it is a continuous process, from gas phase to particle phase. If this is what you mean?

Response: We agree that “condensation” is better here. We have revised as suggested.

Reviewer: Fig.1 The star appears to be red, and not magenta, as it specified in the figure text.

Response: We have changed “magenta” into “red” in the caption of Fig. 1.