Dear Editor,

We would like to thank the two referees for their helpful comments and suggestions, which have been fully taken into account upon manuscript revision. Point-by-point responses to all the comments and a revised manuscript were uploaded.

Best Regards

Chunsheng Zhao

Responses to comments of referee #1

General comments:

1. The manuscript presents measurements and analysis of relative humidity (RH) dependence of aerosol light scattering in a major aerosol source region within NW PRC. The parameterization of the results are of use to the general atmospheric chemistry community and the authors' goal of regionally specific input to global climate models.

Response: Thanks for the comments.

2. Given that the overall HaChi experiment operated from October through January and that this particular subset of the data set is only for 20 days in January, it would be good to put the data subset in context if other data is available. For example, if you have longer term dry nephelometer light scattering, how do the means and standard deviations compare? The point here is to be able to say with some confidence that the hygroscopic and cloud drop nucleating properties that you present are representative of a longer time period or rather are only relevant to the shorter time period – basically a case study. For application to models of radiative forcing, an estimate of representativeness is needed.

Response: Thanks for the comments. To investigate the representativeness of the January data to those obtained from the entire winter campaign, we have made a comparative analysis on aerosol optical parameters observed in the corresponding two periods. The statistical results are shown in the table below, where "**Jan**" and "**All**" represent the period with *f*(RH) measurement and the whole HaChi winter campaign, respectively.

Table 1. Statistical summary of every 5-minute mean aerosol optical properties throughout the campaign (denoted by "**All**"), and during the *f*(RH) measurements in January, 2010 (as displayed by "**Jan**"), respectively.

		Mean		**SD (σ)**		**Median**	
		All	**Jan**	**All**	**Jan**	**All**	**Jan**
σ_{sc} **(Mm^{-1})**	450 nm	552.3	595.1	545.4	545.5	379.4	422.1
	550 nm	448.1	484.5	456.5	458.1	296.0	327.0
	700 nm	310.7	333.5	324.3	322.6	199.5	216.3
σ_{bsc} **(Mm^{-1})**	450 nm	67.4	71.8	62.0	61.7	50.7	55.6
	550 nm	56.3	60.3	52.7	52.7	41.4	45.6
	700 nm	47.1	50.4	45.5	45.4	33.2	36.6
σ_{ab} **(Mm^{-1})**	637 nm	67.1	63.7	51.1	45.8	61.5	54.4
Visibility **(km)**	550 nm	9.4	8.4	10.4	9.1	4.8	4.3

It can be found that the 5-minute mean and median values of both σ_{sc} and σ_{bsc} for the January data subset are slightly higher than those for all the data, while the mean and median σ_{ab} and ***Visibility*** are relatively lower. The corresponding standard deviations (SD) for the both observation periods are comparable with each other. On average, discrepancies between the two kinds of mean values are within 10%, revealing the good representativeness of the January data.

In this study, we mainly focuses on the data analysis during the *f*(RH) observation period. The motivation of this work is to introduce a new retrieval algorithm for the aerosol hygroscopicity parameter *κ* based on the *f*(RH) measurements. As a result, the data obtained in January were used here. To clarify it, we have added more information into the experiment descriptions of section 2.

3. There were two distinctly different synoptic situations during the study in January. I suggest that these be presented as two contrasting case studies. Further, more detail of the meteorology including short term, 3 to 4 day, air mass back trajectories within the boundary layer would be valuable. This would help to define the two events and provide input for eventual input to models.

Response: As stated beforehand, our work is aimed to propose a straightforward method of deriving the aerosol hygroscopicity parameter κ from the f(RH) measurements, rather than to investigate the characterization of f(RH) in the northern part of the NCP region. Taking into account both reviewers' suggestions on the discussion of the two pollution episodes, we have reorganized the paragraphs in the beginning of section 3.1. Figure 2 has been replotted for clearer presentation of the time series of the dry σ_{sc}, wind speeds and wind directions during the f(RH) observation period.

Xu et al. (2011) pointed out that there were two typical synoptic conditions in winter of the NCP region. Specifically, strong winds from the north are often observed with the slightest polluted conditions; while the mild southerly winds would result in severe local pollution due to the accumulation of pollutants with weak diffusion, contributing to the highly frequent haze events. In this sense, we have investigated the variation of aerosol light scattering under different pollution cases determined by the influence of wind parameter. Corresponding discussions have been revised in the manuscript.

Reference:

Xu, W. Y., Zhao, C. S., Ran, L., Deng, Z. Z., Liu, P. F., Ma, N., Lin, W. L., Xu, X. B., Yan, P., He, X., Yu, J., Liang, W. D., and Chen, L. L.: Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in the North China Plain, Atmos. Chem. Phys., 11, 4353-4369, 2011.

4. Several terms are used for the parameter of measure and interest. Stay with one term for readability or maybe two - to not seem monotonous. The general term “Aerosol hygroscopicity parameter” is good and short, for the title. In the abstract “relative humidity (RH) dependence of aerosol light scattering” is a good, properly phrased term, but too long for general use; it can be shortened to “RH dependence of light scattering” for subsequent use. “aerosol light scattering enhancement factor” or later, “scattering enhancement factor“ are fine, too.

Response: We agree with the referee. To the specific term, *f*(RH), we have utilized the following two statements in the context, “RH dependence of light scattering” and “aerosol light scattering enhancement factor”. Correspondingly, we have revised them in the manuscript.

5. The English usage needs to be improved throughout the manuscript. While the science, experiment, results and conclusions are clear to a reader who is familiar with the topic and the literature, it is not easily readable or understandable by the less initiated. Scientific translation is a difficult task, I know, but needs to be done by a co-author or colleague or linguistics professional who is highly fluent in English. I have given some suggestions below but have not worked through the entire manuscript.

Response: We do appreciate the referee’s suggestion and have made corrections throughout the manuscript.

Specific comments:

In the following I have suggested deletions, and additions or changes to text and sentence structure in ~~strikethrough~~ or **bold,** respectively. Or I have simply rewritten the sentence. For my questions or other useful changes I have specified my suggestion or explanations in *italics*.

1. Abstract

On account of the insufficient information of aerosol hygroscopicity in climate models, more details of the parameterized hygroscopic growth factors are urgently required.

Because of ~~the~~ insufficient information ~~of~~ **about** aerosol hygroscopicity in climate models, ~~more details of the~~ **a more detailed parameterization** of hygroscopic growth factors and **resulting optical properties with respect to location, time, sources, aerosol chemistry and meteorology** are urgently required.

Response: Thanks for the suggestion. We have revised it accordingly.

2. Measurements show that *f* (RH) sharply increases with the ascending RH, and the variation range of *f* (RH) is much wider at higher RH.

Measurements show that *f* (RH) increases **sharply** with ~~the ascending~~ increasing RH, and **that** the ~~variation range~~ **time variance** of *f* (RH) is much ~~wider~~ **greater** at higher RH.

This is a more scientific, mathematical wording.

Response: We appreciate the referee for the helpful comment. The corresponding corrections have been done in the manuscript.

3. **A** sensitivity **analysis** reveals …

Response: Thanks. We have revised it.

4. Page 3460

Atmospheric aerosols~~ have exhibited great contribution~~ contribute significantly to….

~~The~~ Aerosol optical properties are crucial input parameters...

Response: We have revised them correspondingly, thanks.

5. Page 3461, line 16

It should be noted that, differ from the size-resolved aerosol diameter growth factor (*g*(RH)), the aerosol light scattering enhancement factor stands for the overall hygroscopicity of the aerosol population, and jointly determined by the particle number size distribution (PNSD), hygroscopicity, and aerosol optical properties.

It should be noted that, differ**ent** from the size-resolved aerosol diameter growth factor (*g*(RH)), *f*(RH)~~the aerosol light scattering enhancement factor stands for~~ **represents** the overall **aerosol light scattering enhancement factor** of the aerosol population, and **is** jointly determined by the particle number size distribution (PNSD), ~~hygroscopicity~~ **chemical composition, density and refractive index** ~~and aerosol optical properties~~.

Response: Thank the referee for the good advice. We have revised it in the manuscript.

6. ~~In the late 1970s,~~ Pilat and Charlson (1966) attempted to measure …

Response: We have corrected it, and thank you.

7. ~~Up to now, the instruments based on the principle of humidified nephelometer measurement have~~ **More recently, the principle of humidified nephelometery has** been improved (Fierz-Schmidhauser et al., 2010a–c). To be specific, ~~with adding a set of PID (Proportional-Integral-Derivative) controller to the humidified~~

~~nephelometer system,~~ **by adding a fast temperature and RH feedback controller to the humidified nephelometer and maintaining a stable reference RH, a quick, automated response of the scanned RH, and *f*(RH) can be achieved.**

Response: Thanks for the comment. We have revised it.

8. Page 3462, line 4

… relatively low RH.

Specify the range Malm used.

Response: Thanks. We have replaced it with "… relatively low RH range of 20-30% RH".

9. **a** comparison results reveal**s** that …

Response: We have revised it. Thanks.

10. It~~'s~~ **is** known that, at a given supersaturation, the aerosol activation ability is ~~primarily~~ determined **primarily** by the particle size **and secondarily by** aerosol hygroscopicity (Seinfeld and Pandis, 1998)

Response: Thanks for the suggestion, and we have corrected it.

11. The online**,** continuous observations of ~~aerosol size~~ **PNSD** can be easily achieved ~~with some~~ **using** commercial instruments. However, ~~the~~ direct measurements of aerosol hygroscopicity are ~~relatively tougher a lot.~~ **relatively much more difficult and demand custom instrumental systems.**

Response: We thank the referee's useful comment, and we have revised it accordingly.

12. In ~~the proposed~~ **our** approach, **we have made** the assumption that the aerosol consisted of simply a soluble fraction of ammonium sulfate and an **unspecified** insoluble component ~~was applied~~.

Response: Thanks for the comment. Actually, the approach here refers to the algorithm proposed by Evans et al. (2007), while our new method is introduced in section 3.4. To avoid ambiguous, we have rewritten the sentence into "In the proposed approach, they have made the assumption that the aerosol consisted of simply a soluble fraction of ammonium sulfate and an unspecified insoluble component.".

13. Page 3463, line 3

I'm not sure what is meant here. Try this.

We show ~~It should be convinced~~ that ~~the~~ **our** retrieval algorithm ~~of~~ **for** *κ* ~~with the~~ **based on** *f*(RH) measurements is of significant utility and applicability.

Response: We rephrased the sentence into "We show that the retrieval algorithm for *κ* based on *f*(RH) measurements is of significant utility and applicability."

14. Recently, with the rapid economic growth, along with the sharp ~~aggravation~~ **expansion** of industrialization and urbanization ~~processes~~, most megacities in the north China plain (NCP) have inevitably experienced severe aerosol pollution. ~~Accompanied~~ **Resulting** aerosol pollution episodes, as well as the aerosol-related environmental and health effects, have aroused great public concern. Considering the unique physical and chemical characteristics of aerosol particles in this region, research~~es~~ on aerosol hygroscopicity **is** ~~are thus~~ of special necessity. Previous studies indicated that aerosols in the highly polluted NCP are ~~of strong~~

~~hygroscopicity~~ **highly hygroscopic** (Liu et al., 2011). Consequently, ~~it would result in~~ **hygroscopic growth of the aerosol will have an** immense impact on aerosol optical **properties** and **cloud droplet** activation~~ properties~~ (Deng et al., 2011; Chen et al., 2012). Nevertheless, due to the limitation**s** of measurement technologies, it~~'s~~ **is** relatively difficult to directly measure ~~the~~ aerosol hygroscopicity, and hence the ~~corresponding~~ **existing** research results are insufficient~~ in this area~~ (Massling et al., 2009; Meier et al., 2009). ~~On account of the observation of~~ **Because measuremen**t of aerosol light scattering enhancement **with integrating nephelometers** is more feasible ~~in~~ **and** practical ~~application~~, many measurements of *f* (RH) have been carried out in the NCP; while relevant studies on *f* (RH) in the northern **part of the** NCP are relatively scarce (Pan et al., 2009; Yan et al., 2009). For better estimation of the radiative forcing by aerosols in the NCP, a comprehensive description of aerosol hygroscopicity and parameterized hygroscopic growth factors are urgently needed in climate models.

Response: We appreciate the good suggestion. Necessary corrections have been made in the corresponding place.

15. Table 2.

Include equation (1) in the caption to make the regression constants more direct and clear to the reader.

Response: Thanks for the suggestion. We have revised it accordingly.

16. *Figure 1.*

Since the relationship is so clear I suggest eliminating the figure and presenting the regression equation, slope and offset in the text. Follow this with your discussion of the regression slope greater than 1.

The TSI nephelometer has minimal electronic or multiple scattering errors at *scattering below 2000Mm^-1. Thus, your second explanation is unlikely the cause of the slope >1. More likely it is due to a difference in RH or in the RH history of the aerosol sample in the nephelometer system along with hysteresis effects.*

Response: Thanks for the constructive comment. Figure 1 has been removed; the regression results and corresponding explanations have been revised in the manuscript.

17. Figure 2.

The time series is difficult to interpret qualitatively or quantitatively. I suggest including it as a supplement to the ACP manuscript.

Response: Considering the two referees' comments, we have replotted Figure 2. To investigate the variation of aerosol light scattering under different pollution levels, along with the wind dependence of σ_{sc}, the modified figure only displays the time series of dry σ_{sc} and wind parameter throughout the f(RH) measurements. Corresponding discussion has been revised in the manuscript.

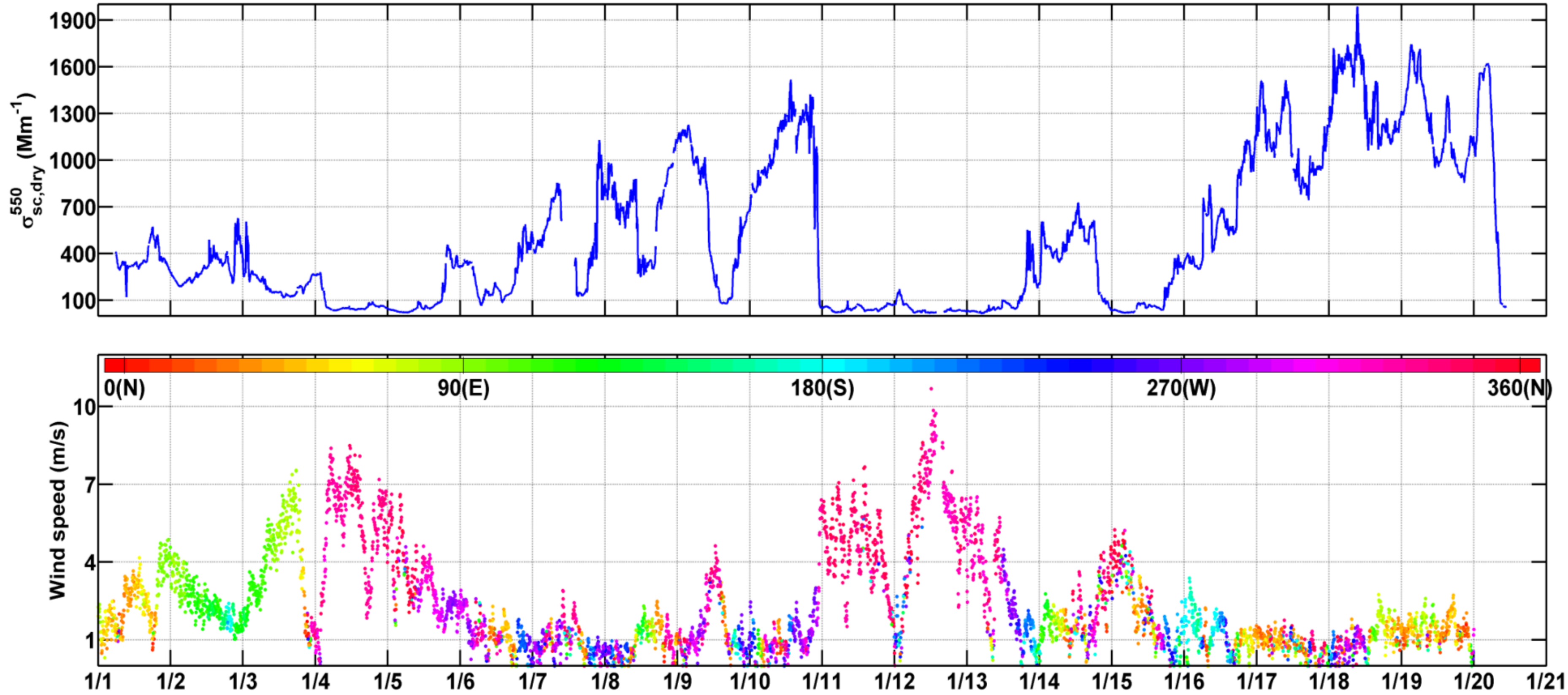

Fig.1 Every 5-minute mean aerosol light scattering coefficients at 550 nm wavelength ($\sigma_{sc,dry}^{550}$) under dry conditions, along with the wind parameter during the *f*(RH) observation periods (colored dots in the lower panel represent the corresponding wind directions).

Responses to comments of referee #2

General comments:

In this manuscript a retrieval method was proposed to calculate the aerosol hygroscopicity parameter *κ*, based on aerosol light scattering enhancement factor *f*(RH) and particle number size distribution during HaCHi campaign. The CCN number concentration was estimated by using the derived *κ*. The method is straightforward and useful to estimate *κ* and to predict CCN concentration. However, several corrections and clarifications are necessary to improve the manuscript. Some discussions in the manuscript need to be better organized. The authors also need to polish the English to void colloquialism. I suggest that the manuscript may be publishable in ACP after revisions listed below.

Response: Thanks for the comments.

Specific comments:

1. In the introduction, it needs to be discussed that particle morphology can also vary considerably with atmospheric aging and RH, impacting the particle optical properties (i.e., Khalizov et al., J. Phys. Chem. 113, 1066, 2009; Pagels et al., Aerosol Sci. Tech. 43, 629, 2009). In particular, how particle morphology variation may impact their algorithm to derive *κ* with the *f*(RH) measurements.

Response: We appreciate the referee for the helpful advice. The corresponding discussion has been added into the introduction section, and the relevant references have been cited. Since the assumption of spherical particles is required for the Mie calculation, the particle morphology changes would cause uncertainty to the *κ* estimation with the *f*(RH) and PNSD measurements. We also have considered the possible influence into the discussion on uncertainty of the derived κ in section 3.4.

2. Page 3463 Line 5-7.

Need to cite several references to discuss the particle pollution in NCP. Need to show numbers, e.g. particle concentrations.

Response: Thanks for the comment. We have added the details in the corresponding place.

3. Page 3463 Line 23-30.

It is unnecessary to describe the contents in every section. Just describe what you have done and why that's important.

Response: Thanks for the suggestion. We have revised it accordingly.

4. Page 3463 Line 23-24.

"…observation…were analyzed"

Response: Thanks. We have revised it.

5. Page 3464 Line 10-11.

Clarify the measurement period. The campaign was conducted from October to January? But the authors only showed the January data.

Response: Thanks for the comment. This work mainly focuses on the data analysis during the *f*(RH) observation period. As what we have introduced in the second paragraph of section 2, the continuous measurement of *f*(RH) was only conducted in January, but not throughout the entire HaChi winter campaign. Therefore, the observations obtained in January were used here. To make it clearer, we have added the sentence below to clarify the motivation of this study.

"This work mainly focuses on the method of deriving κ from the measured f(RH) in January in the northern part of the NCP region."

6. Page 3465 Line 20.

If possible, show the inter-comparison at high RH. As in the discussion afterwards, the CCN comparisons are different at low RH and high RH.

Response: We only have one nephelometer to measure the σ_{sc} under humidified conditions, while the other one operated in parallel is just used to measure the dry σ_{sc}. Thus, the inter-comparison of σ_{sc} at high RH is not available.

As for the different comparison results of N_{CCN} at low supersaturations and high supersaturations, the possible reasons can be concluded briefly as follows. Uncertainties induced by both of the measurement (such as PNSD, σ_{sc}, RH, and N_{CCN}) and calculation (e.g., assumptions applied in the calculation of κ and N_{CCN}) errors would inevitably result in uncertainties of the N_{CCN} comparisons at varying supersaturations. Besides, stable lower supersaturations (SS<0.1%) are relatively much more difficult to maintain as determined by the measurement limitations of the CCNC itself. Hence, the fluctuation of the supersaturation would contribute large uncertainty to the comparison results. Details can be found in section 3.5.

7. Page 3465 Line 18-19.

The Neph measurement should be introduced briefly, not only citing references, e.g how RH is changed during each cycle (Page 3467 Line 8-11 should be moved here), etc.

Response: Thanks. We have revised it accordingly.

8. Page 3465 Line 25.

Explain why weak hygroscopic growth of particles at low RH can lead to high discrepancy at high σ condition.

Response: Taking another reviewer's suggestion into consideration, we have removed Figure 1 from the manuscript, and added the corresponding regression equation, slope and offset in the text. Consequently, we have rewritten the paragraph and discussed the possible causes of the regression slope greater than 1. More information can be found in section 2.

9. Page 3466 the first paragraph.

Re-organize this paragraph. The episodes should be described either by time or types (polluted and clean), e.g., the authors recognize the two pollution episodes, and then descriptions of these two pollution episodes should be made.

Response: Thanks for the suggestion. We have rewritten the paragraphs of section 3.1 as suggested.

10. Page 3467 Line 8-11.

Move this part to the experimental section.

Response: We have revised it accordingly, and thank you.

11. Table1.

List σ for pollution and clean episodes, so that the readers can have the idea of what was the situation in pollution and clean episodes.

Response: Thanks for the suggestion. We have provided the corresponding information in Table 1.

12. Page 3468 second paragraph.

The authors may want to discuss the light extinction, but did not reach any conclusion. If the authors have absorption data or extinction data or visibility, discuss all these data to investigate how much the particle hygroscopicity can affect the visibility.

Response: This work is aimed to develop a straightforward method of deriving the aerosol hygroscopicity parameter κ from the f(RH) measurements. We do not intend to investigate the influence of aerosol hygroscopicity on visibility or light extinction, as which has been discussed comprehensively in our previous work (Chen et al., Atmos. Chem. Phys., 12, 4935, 2012; details can be found in section 4.3).

13. Table 1.

Is the average value for the whole measurement period or just for the clean and pollution episodes? It makes little sense to average only pollution and clean episodes.

Response: We agree with the referee. The "**Average**" f(RH) here represents the overall mean value for the whole f(RH) observation period.

14. Page 3472 Line 11.

"…would be great" colloquialism. There are several English style issues in the manuscript.

Response: Thanks for the comment. We have revised it into "… is evident". Similar corrections have been made where necessary.

15. Section 3.4

Uncertainties in κ estimation should be discussed. The authors discussed the uncertainties in section 3.5, but the discussion should be moved here.

Response: We agree with the referee's suggestion. Discussion of uncertainties in the retrieved κ has been added into section 3.4. We have also revised the content in the corresponding places of section 3.5.

16. Figure 2 is unclear.

Response: Taking into account both referees' comments, we have replotted Figure 2. Time series of the dry σ_{sc} and wind parameter during the f(RH) observation period are presented, aiming for further analysis on the variation of f(RH) under different pollution episodes categorized by the wind dependence of σ_{sc}. Corresponding discussion has been revised in the manuscript.