Referee #2

Received and published: 6 June 2016

Referee Comment

General comment:

The manuscript presents interesting results on aerosol extinction modelling using microphysical aerosol parameters obtained by other measurements. Also, the introduction gives a reasonable overview about the recently published articles in the area. A simple core-shell model is used to compare experimental and modelled data. The dataset is in general a bit short, but sufficient to test the model. Figures and tables are in most cases clear, but figure descriptions need some more work. Especially the paper text is sometimes not fluent and some English correction is recommended before publication. The discussion part is precise, but I suggest to add one or two more figures presenting the results that were used for the model exercises. In general, the Nanjing dataset is used as in a lab study only to test the model. I suggest to put the measure-ACPD ments at this special place into context to similar measurements that were carried out at other sites. The article may be published in ACP when the following comments are taken into account. Interactive

A: First, thank you for your suggestions for this paper. We modified the content carefully according to reviewers' suggestions one by one. We apologize that there were still many sentences with grammatical errors. Thank you for your detailed revision. We have corrected these errors in the amended version and sent the manuscript to AMERICAN JOURNAL EXPERTS (AJE) for further polishing.

Second, the reviewer suggested that we put the measurements from this specific place into context based on similar measurements collected at other sites. We are looking for funding support and plan to make similar observations in Shenyang and Suzhou during the winter. We believe that we will obtain some interesting results.

Detailed scientific comments:

Abstract

Q1: Page 1, Line 15 – 22: Comment: You sometimes use extinction, then extinction coefficient, then volume extinction coefficient. Be clear and specify the parameter you are talking about!

A1: Thank you for your comment. We have modified 'extinction' and 'volume extinction coefficient' to 'extinction coefficient'. (Page 1, Lines17– 23)

1 Introduction

Q2: Page 2, Line 13: Add:...even at subsaturated conditions.

Q3: Page 2, Line 13: Change:...of the particles can lead ...

A2-A3: We have modified the sentence expressions according to the reviewer's advice. 'When the RH is high, even at subsaturated conditions, the hygroscopic growth of the particles can lead to an increase in size…' on Page 2, Lines 16-17

Q4: Page 2, Line 16-17: ...and the extinction associated with different particles is ...Comment: What do you men here with different particles: size classes, chemistry? Please clarify!

A4: We have modified 'different particles ' to 'different particles (with differences in size and chemistry)' on Page2 Lines 20-21.

Q5: Page 2, Line 25: Change: ...of the overall aerosol population.
Q6: Page 2, Line 29: Add: ...based on the observed aerosol.
Q7: Page 3, Line 2: Change: Therefore, we have established ...
A5-A7: Thank you for reviewer's comment. We have modified the sentence expression '…on the physicochemical properties of all particles…' to '…of the overall aerosol population…' on Page 2, Line 29; 'in physicochemical properties of particles' to 'in physicochemical properties of particles based on the observed aerosol ' on Page 3, Line 3; and 'we must establish' to 'we have established ' on Page 3, Line 6.

Q8: Page 3, Line 18: Comment: I would rather more say here that kappa can be considered a function of the volume fraction of the hygroscopic and therewith non-light absorbing components and the volume fraction of the non-hygroscopic and therewith light-absorbing component which here can be assumed to be EC. In this way hygroscopic and optical properties can be understood to have a strong linkage.
A8: Thank you for your comment. The reviewer's phrasing expresses our intended meaning more clearly, so we have adopted the reviewer's expression. We modify 'κ can be considered a function of the chemical composition and volume fraction of the non-light-absorbing component because the hygroscopic growth of EC is poor' to 'κ can be considered a function of the volume fraction of the hygroscopic and therewith non-light absorbing components and the volume fraction of the non-hygroscopic and there with light-absorbing component which here can be assumed to be EC. In this way hygroscopic and optical properties can be understood to have a strong linkage' on Page 3, Lines22-25.

2 Experiment and Methods
Q9: Page 4, Line 18-24: Comment: Which parameter for the size distribution is measured: mass, number? Please specify!
A9: The particle number concentration is used for the size distribution. We have specified as such on Page 5, Lines2-3

Q10: Page 4, Line 26: Change: The size distributions are provided in section as follows:...
A10: We have modified the sentence expression from 'The size distributions were' to 'The size distributions are provided in section as follows ' on Page5,Lines 7-8.

Q11: Page 5, Line 2: Comment: I would expect here a short description (two sentences) of the processing steps.
A11: We have added a short description 'Before use, quartz filters were fires for 5h at 800°C to lower the blank levels for EC and OC. All of these filters were kept in a refrigerator for cryopreservation. Every sample was collected continuously for 23h and then kept in a refrigerator before analyzing (Zou et al., 2014).' on Page5, Line 10-12.

Q12: Page 5, Line 4: Comment: A detailed description of what was provided?
A12: A detailed description of the chromatograph was added on Page5, Lines14-22. 'Chromatography includes the use of a column oven, a conductivity detector, an 858 auto-injector and a MagIC net chromatography workstation (Metrohm, Switzerland). The column oven consists of a Metrosep

C4150/4.0 separation column and MetrosepASupp 5150/4.0 separation column. The eluent was set at3.2mmol • $L^{-1}Na_2CO_3$+1.0mmol • $L^{-1}NaHCO_3$for anions and 1.7mmol • $L^{-1}HNO_3$+0.7mmol • L^{-1} pyridine carboxylic acid forcations. The column temperature was maintained at 30°C. The flow-rate was 1.0mL•min^{-1},and the inject volume was 20 μ L. The detection limits for Na^+, NH_4^+, K^+,Mg^{2+},Ca^{2+}, F^-, Cl^-, NO_2^-, NO_3^- and SO_4^{2-} were0.001, 0.005, 0.001, 0.001, 0.001, 0.01, 0.01, 0.01, 0.01, 0.01mg•L^{-1} respectively(An et al., 2015). '

Q13: Page 5, Line 6: Comment: Again, I would expect here a short description!
A13: A detailed description of the thermal/optical carbon analyzer was added on Page5, Line23-Page6 Line2. 'The samples were heated to 140, 280, 480 and 580°Cin pure He to determine OC1, OC2, OC3 and OC4, respectively. Then the samples were heated to 580, 740 and 840°Cin 2%O_2/98%He to determine EC1, EC2 and EC3, respectively. The volatilized compounds were converted to carbon dioxide (CO_2) through an oxidizer (heated manganese dioxide, MnO_2). CO_2 was reduced to methane (CH_4) through a methanator. Finally, the CH_4 equivalents were quantified with a flame ionization detector (FID). The charring effect can transform part of organic carbon into pyrolysis carbon under anaerobic heating. Hence, the correction for pyrolysis was made by continuously monitoring the filter through a 633nm He-Ne laser. By monitoring the change of reflected light in the heating process, the initial reflected light is an diacritical point of OC and EC'

Q14: Page 5, Line 9-10: Comment: Check this sentence, this is not a full sentence!
A14: We have modified '$PM_{2.5}$ was detected with a β-ray particulate continuous monitor (Thermo Fisher). And the working principles was that measuring particles' mass concentration through β-ray attenuation.' to '$PM_{2.5}$ was detected with a β-ray particulate continuous monitor (Thermo Fisher) with the working principle of measuring the particles' mass concentration through the β-ray attenuation.' on Page6 Lines 2-3.

Q15: Page 5, Line 10-14: Comment: Again, I would expect here a short description of the visibility meter!
A15: We have modified 'The visibility meter was used to measure scattering coefficient of particles and it's light source wavelength was 940nm. A detailed description of these two instruments was provided previously (Yu et al., 2015)' to ' The visibility meter was used to measure the scattering coefficient of the particles and it's light source wavelength was 940nm. The accuracy was ±10% , and the data update rate was 1min. A detailed description of these two instruments was provided previously (Yu et al., 2015) 'on Page6 Lines 6-8.

Q16: Page 5, Line 20-21: Comment: You have to specify which method you used (Gysel or Topping)?
A16: We modified the sentence 'Gysel et al. (2007) used the ion pairing method' to ' We used the ion pairing method from Gysel et al. (2007),....'Page6, Line13-14.

Q17: Page 6, Line 1: Comment: Specify exactly what are the parameters i, N, etc. in formula (1).
A17: We have more precisely specified the parameters in formula (1) on Page6, Lines 24-26. 'where N is the number of pure materials, κ_i is the hygroscopic parameter of the i^{th} pure material, $v_{i,dry}$ is the volume of the i^{th} pure material in the dry condition, and $v_{tol,\ dry}$ is the total volume of the dry particle. '

Q18: Page 6, Line 12-14: Comment: Be correct! Change:...molecular weight of water, ...the ideal gas constant, T the temperature with a value of 20 degree C, ...

A18: We have modified the expression 'molar mass' to 'molecular weight ', 'an ideal gas constant' to 'the ideal gas constant ', and 'T=20°C ' to 'T is the temperature with a value of 20°C ' on Page7,Lines 8-9.

3 Results and Discussion

Q19: Page 6, Line 18: Comment: I would like an explanation here why this simplification of calculation of the extinction coefficient based on visibility measurements can be done. Please give here the scientific background!

A19: We added section2.3, to provide the further scientific back ground on this simplification. An explanation of 3.0/visibility was provided on Page8 Lines21-27.

'The meteorological optical range is determined as (Zhang, 2007):

$$\mathrm{MOR} = \frac{1}{\sigma}\ln\frac{|c|}{\epsilon} = \frac{1}{\sigma}\ln\frac{1}{0.05} = \frac{3.0}{\sigma} \quad (4)$$

where σ is the extinction coefficient of the particles, ε is the visual threshold with a value of 0.05(MOR is equal to the visibility when $\varepsilon = 0.05$), and c is the target characteristic coefficient. When the target is black, c=1.

Hence, the measured extinction coefficient can be calculated from the visibility as:

$$b_{\mathrm{ext,measurement-d}} = \frac{1}{\mathrm{visibility}}\ln\frac{1}{0.05} = \frac{3.0}{\mathrm{visibiliy}} \quad (5)\ '$$

Q20: Page 6, Line 20 Comment: If you use the wording size distribution, you have to specify which parameter you mean. In this case you mean particle number size distribution, I Discussion paper guess. Please take care over the whole manuscript! Also in Figure 3 description.

A20: The size distribution here refers to the number size distribution, and we have modified 'size distribution ' to 'number size distribution ', including the description of Fig3 on Page9 Line6 and Page9 Line 16.

Q21: Page 6, Line 20-22: Comment: Can you give some statistical evidence on this statement. You should look for the correlation coefficient here.

A21: We added a reference to the correlation coefficient on Page9, Line8. '……the periods with high number concentration had a good consistency with periods ofhigh $PM_{2.5}$ mass concentration (r=0.7)' to '……the periods with a high number concentration had a good consistency with the periods of a high $PM_{2.5}$ mass concentration (r=0.7)'

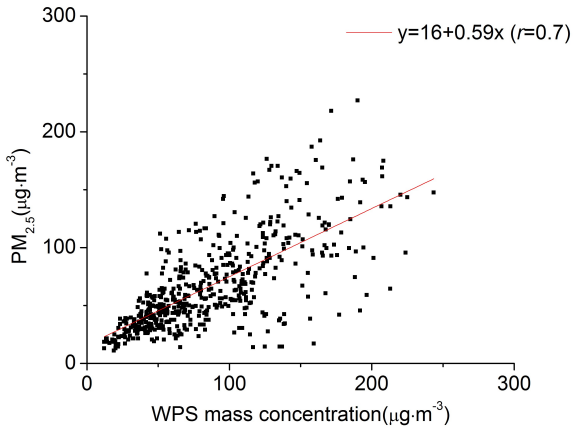

Fig.1 Scatter plot of WPS mass concentration and $PM_{2.5}$ mass concentration

Q22: Page 6, Line 25 and ongoing: Comment: I would say the variation between size segments is higher compared to the variation over time within one size segment especially Interactive for kappa. Please make sure that you mention the time resolution of the Anderson in your study in section 2! I cannot see it there.

A22: First, we have modified the sentence 'but the degree of change in different size segments was larger than that at different times' to 'the variation between size segments is higher compared to the variation over time within one size segment especially interactive for κ' on Page9, Lines12-13. Second, we have given the time resolution of the Anderson as 23h on Page5, Line12. However, the time resolution in the whole discussion part (section 3) is 1h. Page8 Lines1-3 provids an explaination for the time resolutions of the Anderson and WPS.

Q23:Page 7, Line 13 – 15: Please rephrase this sentence. Physicochemical properties can be different for the same size of particles, but in this case the chemical composition is different.

A**23**: We have modified the sentence 'In the real atmosphere, even if the sizes of aerosol particles are the same, the physicochemical properties of particles are significantly different.' to 'The physicochemical properties can be different for the same size of particles, but in this case the chemical composition is different' on Page7 Lines16-17

Q24: Page 7, Line 27 and ongoing: Comment: This section contains some information that should be relocated to the instrumental section 2 or even is there now. Please shorten and make more clear!

A24: We have relocated some of the information in section 3.2 to section 2.3.

Q25: Page 8, Line 12: Add: ...of the extinction coefficient...

A25:We have modified 'Figure 6 shows the relative values of the calculated and observed values from the core-shell model' to 'Figure 6 shows the relative values of the model and measurement values of the extinction coefficient from the core-shell model 'on Page9,Lines22-23.

Q26: Page 8, Line 20: Change: ...the following section focusses on the measurements and calculations at lambda = 550 nm for discussion.

A26: We have modified the sentence 'the following section adopts 550 nm for discussion' to 'the

following section focuses on the measurements and calculations at λ=550 nm for discussion.'on Page10, Lines1-2

Q27: Page 9, Line 1: Comment: I would rather call this the relative contributing fraction of different size segments to the dry aerosol extinction coefficient. The same I suggest for Figure 7 and Figure 8 descriptions and all other parts of the manuscript.

A27: Thank you for reviewer's advice. We have modified 'of the extinction coefficients in different size segments under dry conditions' to 'of different size segments to the dry aerosol extinction coefficient' on Page 10 Line11, 'contribution fractions of the extinction coefficients in different size segments under dry conditions' to 'relative contributing fraction of different size segments to the dry aerosol extinction coefficient' on Page 10 Line11-12, 'of extinction coefficients in different size segments at ambient RH ' to 'of different size segments to the wet aerosol extinction coefficient' on Page11 Line3, 'contribution fractions of the extinction coefficients in different size segments at ambient RH' to 'relative contributing fraction of different size segments to the wet aerosol extinction coefficient ' on Page11 Line4-5. 'in different size segments at ambient RH ' to 'of different size segments to the wet condition' on Page11 Line6, 'contribution fractions of the extinction coefficients in different size segments ' to 'relative contributing fraction of different size segments to the aerosol extinction coefficient' on Page11 Line7. And the same for Figure 7, Figure 8 descriptions on Page10 Line24-27.

Q28:Page 9, Line 3 and ongoing: Comment: This is really an interesting finding. But I still understand that you measured PM2.5 and not PM10! I suggest here that a plot showing the overview of particle number or even better particle mass size distribution is included, maybe only as an average value for the whole campaign.

A28: In our study, the PM_{10} mass concentration was calculated by WPS data. We added Fig.9 to Page10 Line28.'Extinction capacity relative to unit mass in different size segments under dry/wet conditions'. and a description of this figure on Page10 Lines 18-24. 'To verify this point, we present Fig.9, which reflects the extinction capacity relative to the unit mass in different size segments under dry/wet conditions. The y-axis is the ratio of the extinction coefficient to the mass concentration for different size segments. From the picture, we can find that extinction capacity relative to the unit mass in the 0.2-2 μ m range was much stronger than that of the other segments. This result explaines why the particles in the 0.2-2 μ m range are the most important for the reduction of the visibility, especially those in the 0.5-1 μ m range.'

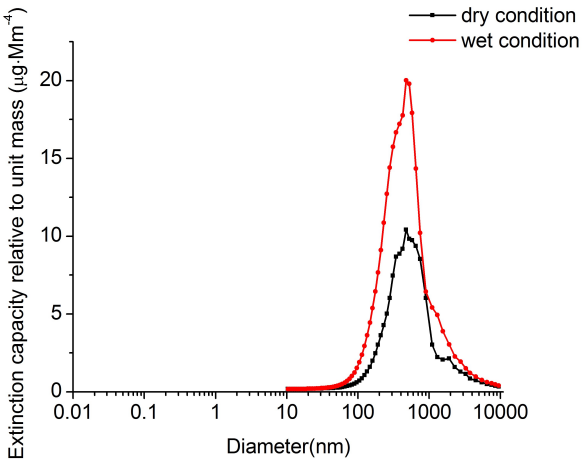

Fig.2 Extinction capacity relative to unit mass in different size segments under dry/wet condition

Q29: Page 9, Figure 9: Comment: Figure axis units and legends/symbols are too small. Figure description is incomplete as missing for Figure 9b.

A29: First, because we added a picture before Fig9, the original Fig.9 has become Fig.10. We have increased the font size in Fig.10, and performed the same process on all the other pictures throughout the manuscript. We also modified the title of Figure 10a/10b on Page12 Lines 24-25. 'Growth multiples of the extinction coefficients（a）and the change in the efficiency factor (b) for different size segments at ambient relative humidity '

Q30:Page 9, Line 13 – 22: Comment: Put the absolute measurements in context to data from other sites you will find in the literature! Otherwise, you only use Nanjing as a lab study to test your model.

A30: In the literature, we usually find that the particles in the 0.2-2 μ m range correlate well with the visibility. However, with the RH increasing, depth analyse of extinction in different size segments are rare. In this study, we take Nanjing as an example to perform a test and depth analysis, providing the explanation that the particles in the 0.2-2 μ m range correlate well with the visibility. Now, we are looking for funding support and plan to perform observations in Shenyang and Suzhou during the winter. We believethat this study will obtain some interesting results..

Q31:Page 9, Line 23 – Page 10, Line 11: What can you conclude from this finding. Can you look into the chemistry and find possible explanations!

Q31:Fig10b provides the explanation for this part. We swapped information on Page12 Lines15-23('Because the average particle size distribution…0.5-1.0μm and 2.0-10.0μm size ranges decreased slightly.') with information on Page11 Line24-Page12 Line14 ('The impact of RH on particles was reflected…extinction coefficients vary at different RH levels')

Q32: Page 10, Line 12 – Page 11, Line 1: Comment: Honestly, I got lost in this paragraph. comment Please choose a more appropriate way to explain the details of your calculations and corresponding findings!

A32: Please see response to Q31. We swapped the information of Fig. 10b with that of Fig. 11.

4 Conclusions

Q33: Page 11, Line 10: Add:...measured values (...) that were derived from... Detailed technical and language comments

A33: We added '…that were derived from the visibility…' on Page13 Line6.

Q34-Q58 are correction of sentences and grammatical errors. Thank you for reviewer's suggestions. We have modified these carefully, and send this paper to AMERICAN JOURNAL EXPERTS(AJE) to polish the full text again.

Abstract

Q34: Page 1, Line 15: Change:...produced reasonable results.

A34: We have modified '…was reasonable 'to '…produced reasonable results' on Page1 Line 17.

1 Introduction

Q35: Page 2, Line 3-4: Change: ...is mainly caused by the increase of particle number or mass concentrations and can lead...(...such as cardiovascular diseases:::) and can further lead to an increase of traffic accidents ...

A35: Thank you for your advice. We have modified 'Visibility degradation mainly caused by the increase of particle concentration and easily lead to a variety of health problems (such as cardiovascular disease, respiratory system diseases, etc.) and traffic accidents increasing' to 'Visibility degradation is mainly caused by the increase of particle number or mass concentration and can lead to a variety of health problems (such as cardiovascular disease, respiratory system diseases, etc.) and can further lead to an increase of traffic accidents,' on Page2 Lines5-9.

Q36: Page 2, Line 28: Change ...though...

A36:We have modified "thought" to "though" on Page3,Line2

Q37: Page 3, Line 9: Change: ...indices...

A37: We have modified "indexes" to "indices" on Page3, Line13

Q38: Page 3, Line 11: Change: She found ...

A38: We have modified "He" to "She" on Page3, Line16

Q39: Page 3, Line 15: Comment: There are two points after the sentence.

A39: We have deleted one point here. (Page3 Line20)

Q40: Page 3, Line 21: Change: ...of the particles:::Then, we can calculate...

A40: Thank you for reviewer's advice. We have modified 'we can determine the changes in volume of both the real part and imaginary parts, then calculate the extinction coefficient of particles ' to 'we can determine the changes in volume of both the real part and imaginary parts of the particles, then we can calculate the extinction coefficient of particles ' on Page3 Lines26-28

Q41: Page 3, Line 24: Add: ...of the observed aerosols.

A41: We have added '...of the observed aerosols' on Page4 Line1.

Q42: Page 4, Line 3: Change: ...in dependence of RH and ...different particle size ranges.

A42: We have modified 'the extinction coefficient as the RH increased ' to 'the extinction coefficient in dependence of RH ' on Page4, Line11.

2 Experiment and Methods

Q43: Page 4, Line 8: Shorten:...above ground level.

A43: We have modified '40 m above the ground ' to '40 m aboveground level' on Page4 Line16.

Q44: Page 5, Line 6: Change: EC and OC were determined ...The measurement principle...

A44: We have added a detailed description of the thermal/optical carbon analyzer according, as stated in the response to Q13, on Page5, Line23-Page6 Line2. The sentence mentioned by the reviewer has been deleted.

Q45: Page 5, Line 25-26: Change: We obtained ...density of 1.7...we calculated...

A45: We have modified 'We can obtain the mass of each pure species according to the pairing method. Supposing a dry particle's density is 1.7 $g \cdot cm^{-3}$(Wehner et al., 2008), we can calculate the volume of the dry particle.' to 'We obtained the mass of each pure species according to the pairing method. Supposing a dry particle's density of 1.7 $g \cdot cm^{-3}$(Wehner et al., 2008), we calculated the volume of the dry particle. ' on Page6 Lines19-20.

Q46: Page 6, Line 3: Add:...material component

A46: We have added 'component' on Page6 Line27.

3 Results and Discussion

Q47: Figure 1: Change: Data coverage...

A47: We have modified 'Missing data from instruments…' to 'Data coverage from instruments…' on Page4 Line22.

Q48: Figure 2: Change:...observation period.

A48: We have added 'period' on Page20 Line6.

Q49: Figure 3: Change ...particle number size distribution ...

A49: We have modified 'Time series of size distribution (dry particles)…' to 'Time series of particle number size distribution (dry particles)…' on Page9 Line16.

Q50: Page 6, Line 21 Change: ...that the periods with high number concentrations had a good consistency with ...

A50:We have modified the sentence 'the periods of high particles number concentration had a good consistent with periods ofhigh $PM_{2.5}$ mass concentration ' to 'the periods with a high number concentration had a good consistency with the periods of a high $PM_{2.5}$ mass concentration (r=0.7)' on Page9 Line7-8.

Q51: Page 6, Line 20-22: Change: ...for different particle size segments...

A51: We have modified 'for different particle sizes' to 'for different particle size segments ' on Page9 Line9.

Q52: Figure 4: Change :...in different size segments ...
A52: We have modified 'Time series of κ for different sizes…' to 'Time series of κ in different size segments…' on Page9 Line17.

Q53: Page 6, Line 20-22: Change: ...for different particle size segments...
A53: We have modified 'for different particle sizes' to 'for different particle size segments ' on Page9 Line9. (same as Q51)

Q54: Page 7, Line 18: Change:...the calculated results...
A54: We have modified '…the calculation results ' to 'The calculated results...' on Page7 Line20.

Q55: Page 7, Line 24 – 25: Change: ...GF is a function of kappa and the hygroscopic uptake of EC is minor...
A55: We have modified 'GF as a function of κ, the hygroscopic level of EC is poor' to 'GF is a function of κ and the hygroscopic uptake of EC is minor…' on Page7 Lines26-27.

Q56: Page 8, Line 23: Change: Contributing...
A56: We have modified the sentence on Page10 Line5.

Q57: Page 9, Figure 26: Change: ...was larger than ::.
A57: We have modified '…was more than…' to '...was larger than…' on Page11 Line18.

4 Conclusions
Q58: Page 11, Line 24 Change:...size ranges.
A58: We have modified '2.0-10.0μm ranges' to '2.0-10.0μmsize ranges ' on Page13 Line21.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-119, 2016.