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Atmospheric
Chemistry
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Discussions

Interactive comment on "Parameterization of cloud droplet formation for global and regional models: including adsorption activation from insoluble CCN" *by* P. Kumar et al.

P. Kumar et al.

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We thank the reviewer for the very thorough, extensive, thoughtful and constructive comments.

General comments

P. Kumar et al. present a new cloud droplet activation parameterization. . . I would like to see an important addition to the presented work: It would be illuminating to see which percentage of the FHH particles gets activated for the simulations presented in section 4.4.1 and 5., and to which extend this depends on the fraction of Köhler particles with which they are competing for water vapor. Adding such an analysis would help the reader to assess how important adsorption ac-

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tivation possibly is in the atmosphere and whether it is necessary to include it into global, regional or cloud models. Of course such a conclusion would probably depend on the choice of the adsorption parameters, but it would add a lot of practical relevance to this otherwise rather theoretical (and hypothetical) paper.

This is a good point. A thorough analysis of course requires a detailed study that would greatly expand the size of an (already large) paper, but we have included one sensitivity study to illustrate the effect of FHH particles on the activation fraction of Köhler particles in the revised manuscript.

Detailed comments

p. 16853: I suggest to speak of "cloud albedo effect" and "cloud lifetime effect", rather than first and second indirect effects, because these names are more meaningful and also endorsed by IPCC.

Done. Names changes as suggested.

Please give a reference (can be a textbook) for the FHH isotherm on top of p. 16855.

Done.

p. 16855, l. 10: I would be curious to know why you prefer FHH theory over BET?

A number of reasons were taken into consideration when developing FHH theory parameterization over BET theory:

- FHH theory is a two parameter fit model (A_{FHH} and B_{FHH}) as opposed to one parameter (c) for BET theory.
- FHH theory makes use of the fact that first layer as well as subsequent layers interact with the solid surface. But in BET theory, only first layer interacts with the solid surface; subsequent layers have the properties of bulk water.

- Sorjamaa and Laaksonen, 2007 suggest that FHH theory distinguishes the adsorption behavior at high RH's on different types of surfaces better that the BET theory.
- Also at RH's very close to 100%, Sorjamaa and Laaksonen, 2007 indicate that the BET isotherms become indistinguishable after the water coverage is a few tens of monolayers where as FHH isotherms converge at much higher coverages.

p. 16855, l. 10: Sorjamaa and Laaksonen (2007) state that their results are only applicable to "perfectly wettable insoluble particles". Please include this restriction somewhere.

Done.

p. 16855, l. 21: As Θ is commonly often used as symbol for a fractional surface coverage ($<$ 1), it would be good if you could add here as a description "number of monolayers".

Good point. Done.

It would be nice to show a plot of eq. (4) e. g. for a stable and an unstable case.

Good point. Done and presented as Figure 1.

p. 16860, l. 15: Please explain where $x = -3/2$ comes from.

For Köhler particles, the relationship between critical supersaturation, s_c of a particle and dry diameter D_{dry} is given by $s_c = CD_{dry}^{-3/2}$ (Fountoukis and Nenes, 2005) hence $x = -3/2$

eq. (14): mathematically seen, the ad hoc introduction of the minus sign is wrong. Please rewrite such that this makes sense.

The mathematical Eq. (14) is correct as it follows chain rule of derivatives. In$D_{dry}$is normalized by dividing it by 1 μm otherwise InD_{dry} is not defined. The term $\frac{d\ln D_{dry}}{ds}$ is inherently negative. The introduction of negative sign to make the right hand side of Eq. (14) positive, hence making $\frac{dN}{ds} > 0$.

p. 16861, l. 1: $dDp = -ds$ is mathematically seen wrong, although I understand what you mean. Please write a whole sentence.

Thank you for pointing this out. It is now corrected, written as words.

p. 16862, l. 4: "wide range of $D_{g,i}$ (0.03-0.1μm)": Did you really use the whole range? Figure 2 shows only two values of $D_{g,i}$.

Yes, we did, but it is impractical to provide plots for the entire range of values considered.

p. 16862, l. 4: Why did you not go higher up in diameter? Later you use diameters up to 10 μm. Is your fit still applicable to the higher values?

Good point! Yes, the fit is applicable at higher diameters equal to 10 μm., and is now shown.

p. 16862, l. 4: Table 3 lists 10 combinations of A_{FHH} and B_{FHH}. Why did you use only 8 here?

Good point. We now provide results for all 10 combinations of A_{FHH} and B_{FHH} mentioned in Table 3.

p. 16862, l. 7: It is not clear to me whether x is supposed to be independent of D_{dry}. Are the slopes the same for Fig. 2 (a) and (b)? Are they still the same for much larger values of D_{dry}? For which values of $D_{g,i}$ and D_{dry} did you calculate the data shown in Fig. 3?

Yes. x is independent of D_{dry}. The discussion clearly states that x is a function of only A_{FHH} and B_{FHH}. The slopes are same for the same combinations of A_{FHH} and B_{FHH} in Fig. 2(a) and (b). The slopes are same for higher values of diameters. We provide an additional plot for $D_{g,i}$ = 10 μm. We have stated the ranges of D_{dry} and D_{gi}

used to produce Fig. 3. $D_{dry}/D_{g,i}$ varies accordingly as illustrated on the x – axis of Fig. 2.

p. 16862, eq. (20) and (21): It is a bit confusing whether i and j are indices or exponents, please state explicitly. Why did you chose a polynom of the order (-3,-4)?

We have clarified this in the text. We have also repeated the fitting with another function that better reproduced the data, over the complete range of FHH parameters considered.

Fig. 3: Are the lines connecting the points given by the fit parameters? Or is this a higher order fit produced by the plotting programm? Please state in the figure caption. It actually looks weird that e.g. the bright green line should peak at $B_{FHH}\sim$ 1.3; have you tested that it does not go further up for $B_{FHH} <$ 1.2? This range can be quite important as it has been suggested that B_{FHH} is close to 1 for black carbon. And what about values of $B_{FHH} <$ 1, which also have been observed? Is your fit still applicable for them? The calculation of x is a crucial step in the derivation of your parameterization, so you should give more details here, and revise Fig. 3 for a larger range of B_{FHH}, more data points for B_{FHH} around 1, and compare the fit to the data points in the figure (or add in the caption that this is already done).

All these are excellent points. We have remade the Figure, and now symbols represent the values of x calculated from adsorption activation theory, and the lines represent the fit (Eq. 20). We have tested the validity of the fit for $B_{FHH} <$ 1.2 and extended the graph to cover the entire B_{FHH} . We have also added more points around $B_{FHH} = 1.0$.

p. 16863: It is rather unfortunate that you define α differently from Barahona and Nenes (2007), that makes it difficult to follow your calculations. But it seems that ρ_a from the denominator of the first term from Barahona and Nenes, eq. (13), is missing here. Please check.

$\alpha$is defined same as Barahona and Nenes (2007) for the simplified representation. Please see paragraph [13] in Barahona and Nenes (2007). Thank you for pointing out the typo in Eq. (24) was a typo. It is now corrected.

p. 16863, l. 8: τ is not explained.

Now explained.

p. 16863, l. 18: The following equations are also valid for e $\neq$ 0, as you defined α to include the entrainment term. Do you exclude entrainment from here on? Please specify.

We do not exclude entrainment from this point onwards in the development of our framework.

We set the entrainment term e equal to 0 to test for different activation physics. This is specified in p. 16863 l 18.

p. 16863, l. 18: Why does $I_e(0, s_{max})$ have the index e? It does not depend on e.

We follow the nomenclature of Barahona and Nenes (2007), where Eq. (14a) (Barahona and Nenes, 2007) can be simplified to Eq. (14c) (Barahona and Nenes, 2007). Here we test the parameterization by equating $e = 0$, but the parameterization would work equally well for $e \neq 0$, and justifies the usage of integral with subscript e.

p. 16864, l. 12: "where D_v' is ..." should read D_v.

Thank you for pointing out the typo. Now corrected.

p. 16865, l. 10: Which unit has $D_{p,low}$?

The units are μm. Now specified.

p. 16866, l. 8: Why 1500? 50^3 = 125000.

Thank you for pointing out the typo. Now corrected.

p. 16866, l. 9: “This means that $D_p >> D_c$ can be assumed for all FHH particles.” I do not understand how you come to that conclusion. The values in Table 1 do not say anything yet about how quickly D_c is reached, or which supersaturation is required to reach it. Does this mean that you assume that all FHH particles become activated before the first Köhler particles get activated? This does not make sense. Sorjamaa and Laaksonen (2007) show that this is only the case for subset of (A_{FHH}, B_{FHH}) values, and state that it is not clear whether these values are realistic. Therefore this should not be your standard assumption.

Table 1 shows that for a FHH particle of given dry diameter, D_{dry}, its critical diameter D_c, is much lower than if it were a Köhler particle (this comparison has been done assuming that particles are composed of $(NH_4)_2SO_4)$. This still applies if the comparison is done between FHH and Köhler particles of the same critical supersaturation (but for brevity is not shown). We have rewritten this whole section to reflect this and to clarify the conditions for which the assumption of $D_c/D_{dry} < 2$ applies.

We never said that FHH particle will activate “before” any Köhler particle, but that it requires substantially less water to attain its critical diameter (and become unstable) *after* it is exposed to $s > s_c$. Whether FHH or Kohler particles activates “first” (in a Lagrangian sense) depends solely on their CCN spectra, $F^s(s)$.

p. 16866, eq. (32): Nenes and Seinfeld (2003) have an additional factor G in their corresponding equation (22). Is this a typo or can you explain it?

Thank you for pointing out the typo. Now corrected.

p. 16866, eq. (32): Please use another letter than x here, as x has a different meaning e.g. in eq. (33).

Done. Now changed from “x” to “y”.

p. 16867, eq. (34): I think you have a factor Gs_{max} too much here (it is already contained in the I terms).

Thank you for pointing out the typo. Now corrected.

p. 16868, l. 13ff: There are several inconsistencies between the text and Table 3. First, Table 3 gives only 150 combinations of the parameters, not 6400. Second, does A_{FHH} start at 0.5 or 0.25? Does B_{FHH} go up to 2 or 2.5? 290 or 298K? The pressure of 1.013 kPa must be a typo, I assume. Table 3 says 900000 Pa, which is equally absurd, but in the opposite direction.

Table 3 gives 150 combinations however we performed a total of 6600 simulations for comparisons using the distributions mentioned in Table 4 and Table 5. For each aerosol distribution we considered an external mixture of Köhler and FHH particles, allowing the proportion to go from 100% Köhler and 0% FHH, to 0% Köhler and 100% FHH (by number).

All typos are now corrected.

p. 16868, l. 13ff: Why don't you span the whole range of observed values of A_{FHH} and B_{FHH} (as given on p. 16857, l. 13)?

We span the entire range of A_{FHH} and B_{FHH} as shown in Fig. 1 and select combination of A_{FHH} and B_{FHH} which do not lie in Region 1 (i.e. those that converge to give s_c and D_c).

p. 16868, l. 25: What about the 50/50 partitioning for soluble/insoluble mass? If all particles are internally mixed, shouldn't they all be Köhler particles? Or do you assume this only for the Köhler particles? If you assume Köhler particles with an insoluble core, the relevant equations (7 etc) would have to be modified. I think it would be good if you list the Köhler particle properties also in Table 3, even if you keep them constant.

We consider a parcel of externally mixed FHH and Köhler particles. The particles are not internally mixed. Köhler particles are 50% soluble (with properties of $(NH_4)_2SO_4$), while FHH particles are insoluble with adsorption properties expressed by A_{FHH} and

B_{FHH}. These properties are now listed in Table 2. In computing s_c for Köhler particles the soluble fraction is accounted for in the n_s term of Eq.(7). All these points are not introduced in the text.

p. 16869, l. 7: Please comment on whether the agreement is equally good for a different choice of parameters.

Yes, the agreement works equally well for different choice of parameters (size distributions, accommodation coefficients, updrafts, Köhler and FHH parameters). We provide additional plot for Whitby distributions, A_{FHH}= 2.00, B_{FHH} = 1.00 and α_c = 0.06 with R^2 value = 0.998.

sections 4.3 and 4.4: Do all your results plotted here contain varying fractions of FHH particles? It would be necessary to state this again here.

Yes. We mention this explicitly at the end of section 4.2 “Evaluation of involved parameters” which implies that for each aerosol size distribution (Sect. 4.3 and Sect. 4.4) we consider an externally mixed parcel of Köhler and FHH particles, allowing the proportion to vary from 0% (pure Köhler particles) to 100% (pure FHH particles). We have restated this for completeness.

section 4.4.1: Why are the data for urban aerosol much more clustered than for the other aerosol specifications, which vary more smoothly?

This is due to the shape of the size distribution; as a result, the slope of the cumulative CCN spectrum is relatively steep at low s, flattens out at intermediate s, steepens again at high s before it flattens out to zero at high particle sizes. The data points are clustered because they correspond to “intermediate” supersaturations. The other CCN spectra are much more like classical “sigmoids”, and are much more distributed in the figure.

p. 16869, l. 16: You give the mean error, which is very small. What is the root mean square error? I think this would be a more meaningful variable to look at.

(Also at other locations in the text.) I do not understand how the 0.37% given here relates to the values given in Table 6.

RMS error gives a measure of the absolute error. Relative error gives a measure of the bias. 0.37% indicates there is no systematic bias. Standard deviation gives a measure of the scatter and is used in place of RMS. Based on this, we can say that approximately 70% of parameterization calculations agree with the parcel model to within 20%. As discussed in section 4.4.1, the average relative error of 0.37%±16% is specific to the data presented in Fig. 7. Table 6 provides results for both Whitby and Dust representative distributions for all values of conditions considered as mentioned in Table 3.

section 4.4.1: Do you have any explanation why the agreement is so much worse for the mineral dust distributions than for the Whitby distributions? Is it that the parameterization in general has problems with large particles?

We provide an explanation for this in Sect. 4.4.1 and Sect. 5 (now revised Sect 4.4.2). Please refer to P 16869, line 19-26 and P 16870, line 22-25. This problem could be corrected if the dry size of the particle is included in the condensation integral, but is left to be addressed in a future study.

p. 16875, Table 1, footnote b: You give M_w twice. Please specify which substance you assume. Is this the same for which the values (0.67, 0.93) have been measured?

Typo. The second one is $M_{(NH4)2SO4}$ = 132.14 g mol^{-1}. The values of A_{FHH} and B_{FHH} cited (0.67, 0.93) correspond to adsorption of water upon $(NH_4)_2SO_4$ (Sorjamaa and Laaksonen, 2007), hence the comparison is for the same substance.

Table 6: You never refer to the last three lines of this table, could be added to section 4.4.2. Why is C04 missing here?

Thank you. We will include discussion in Sect. 4.4.2. We have also included C04 in

Table 6.

Technical comments

p. 16856, l. 8: I would not call Θ a “scale” because it is dimensionless.

We meant “scaled parameter”. Now corrected.

p. 16857, l. 21/22: “lies between 1-2, which suggests that D_c is very close to D_{dry}”: a factor 2 is not very close. Please reformulate.

The statement should be viewed within the context of comparing against D_p of droplets at the point of maximum supersaturation. We have now clarified this in the text.

p. 16864, l. 9 versus p. 16863, l. 16: Be consistent with D’v or Dv’ (prime in index or not).

Corrected.

p. 16868, l. 1: at - $>$ et

Corrected

Section 5 should better be section 4.4.2.

Changed.

Numerous authors names of the papers which you cite are mispelled. This nearly comes across as a lack of respect versus your colleagues. E.g. your most important reference, “Sorjamaa and Laaksonen”, is mispelled throughout the text. Also “Frenkel”, “Heymsfield”, “Wiegner”. The Feingold and Heymsfield paper is in JAS, not JGR. Please check the rest of the references very carefully for more errors.

We truly apologize for these oversights and have made appropriate changes in the text to rightly address all the authors.

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Cite rather the appropriate chapter of the IPCC report (with the author list as given on the first page of that chapter) than “IPCC, 2007”.

Included.

The reference “Asa-Awuku and Nenes (2007)” (p. 16864, l. 16) is missing.

Added.

Table 1 should come after Table 2.

We have reformulated section 2.2 such that Table 1 comes before Table 2.

p. 16870, l. 11: right down - > right panels

Removed.

p. 16875, Table 1, footnote a: Change A_ to Å.

Done

p. 16875, Table 1, footnote b: Change (8) to (8b).

Done

Fig. 4: It would be more reader-friendly to colorcode the lines in the same way as in Figure 3.

Changed.

Fig. 8: It would be better to have a legend with the color symbols instead of the black- and white version in the caption, as e.g. the - and + symbols are hardly distinguishable.

Changed.

Fig. 10: The axis titles of the individual plots have different font sizes.

Fixed.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16851, 2008.

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