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Atmospheric
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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.

Anonymous Referee #3

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General comments

P. Kumar et al. present a new cloud droplet activation parameterization which is not only based on Köhler theory, but includes the possible activation of particles via adsorption of water molecules on wettable surfaces. This work is innovative, highly relevant for studies of the aerosol indirect effect, and it merits publication in ACP. However, I have a major concern on the derivation of eq. (31), see comment below, although I admit that I might be misunderstanding something here. Connected to this, I would like to see an important addition to the presented work: It would be illuminating to see which

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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 activation 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.

The paper is generally well written, but there were quite a number of mistakes and inconsistencies, see below.

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.
- Please give a reference (can be a textbook) for the FHH isotherm on top of p. 16855.
- p. 16855, l. 10: I would be curious to know why you prefer FHH theory over BET?
- 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.
- 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”.

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- It would be nice to show a plot of eq. (4) e. g. for a stable and an unstable case.
- p. 16860, l. 15: Please explain where $x = -3/2$ comes from.
- eq. (14): mathematically seen, the ad hoc introduction of the minus sign is wrong. Please rewrite such that this makes sense.
- p. 16861, l. 1: $dD_p = -ds$ is mathematically seen wrong, although I understand what you mean. Please write a whole sentence.
- 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 $D_{g,i}$.
- 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?
- p. 16862, l. 4: Table 3 lists 10 combinations of A_{FHH} and B_{FHH}. Why did you use only 8 here?
- 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?
- 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)?
- 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 $B_{FHH} \approx 1.3$; have you tested that it does not go further up for $B_{FHH} < 1.2$?

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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, more data points for B around 1, and compare the fit to the data points in the figure (or add in the caption that this is already done).

- 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.
- p. 16863, l. 8 ff: τ is not 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.
- p. 16863, l. 18: Why does $I_e(0, s_{max})$ have the index e? It does not depend on e.
- p. 16864, l. 12: “where D_v' is ...” should read D_v.
- p. 16865, l. 10: Which unit has $D_{p,low}$?
- p. 16866, l. 8: Why 1500? $50^3 = 125000$.
- 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

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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.

- 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?
- p. 16866, eq. (32): Please use another letter than x here, as x has a different meaning e.g. in eq. (33).
- p. 16867, eq. (34): I think you have a factor Gs_{max} too much here (it is already contained in the I terms).
- 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.
- 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)?
- 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.
- p. 16869, l. 7: Please comment on whether the agreement is equally good for a different choice of parameters.

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

Technical comments

- p. 16856, l. 8: I would not call Θ a “scale” because it is dimensionless.
- 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.
- p. 16864, l. 9 versus p. 16863, l. 16: Be consistent with D_v' or $D_{v'}$ (prime in index or not).

- p. 16868, l. 1: at – > et
- Section 5 should better be section 4.4.2.
- 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.
- 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”.
- The reference “Asa-Awuku and Nenes (2007)” (p. 16864, l. 16) is missing.
- Table 1 should come after Table 2.
- p. 16870, l. 11: right down – > right panels
- p. 16875, Table 1, footnote a: Change A° to Å.
- p. 16875, Table 1, footnote b: Change (8) to (8b).
- Fig. 4: It would be more reader-friendly to colorcode the lines in the same way as in Figure 3.
- 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.
- Fig. 10: The axis titles of the individual plots have different font sizes.

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