Comments to the manuscript with MS No. gmd-2020-404

General comments:

1. The authors have improved the presentation of the manuscript substantially in this revision. However, it is still hard to read. It took me more time to understand and comment on the writing instead of science.

I suggest all the authors read through the manuscript carefully to improve the readability. I would also encourage the authors to explain each figure clearly and logically. Please see my suggestions in the section of specific comments.

2. It is still unclear to me what the advantage of using the upwind scheme over high-order schemes is. How is the computational cost of this scheme compared to the high-order schemes? Discussing this briefly in the introduction and the discussion part can be beneficial for the community.

Indeed, in the result section, the authors argue that "... As a result, the algorithm is characterised by reduced numerical diffusion while maintaining the salient features of the underly ...". Which benchmark scheme did you compare with?

3. The authors found that "... even a tenfold decrease of the spurious numerical spectral broadening can be obtained by an apt choice of the MPDATA variant (maintaining the same spatial and temporal resolution), yet at an increased computational cost ...". In my opinion, this is an interesting finding since it helps understanding the artificial broadening of particle-size distribution. What is the benchmark for this conclusion?

I am not asking for performing more simulations. Comparing your current results to some references should be enough to address the question.

4. Is the "box model simulation" direct numerical simulation? What is your numerical setup? Is it a 1-D case driven by a constant supersaturation? What are the governing equations of the motion of particles? How do you determine your time step? What are the boundary conditions?

5. What is the link between the "box model simulation" and the single-column model? Shouldn't the results from "box model simulation" be used or compared to the sub-grid scale modeling of large-eddy simulations before jumping to the single-column model? Can the conclusion of MPDATA from the box simulation be carried over to the single-column simulation? I encourage the authors to discuss this link in depth, which can be a highlight of this study.

Again, I **cannot** recommend the publication in its current form. A major revision is required for further consideration.

Specific comments:

1. P1, L5: Could you please rewrite "The numerical diffusion problem inherent to the employment of the fixed-bin discretisation in the numerical solution of the arising transport problem is scrutinised." to make it more concise?

2. P1, L5: What is "carried out"? Neither "Eulerian modelling approach" nor "evolution of the probability ..." fit the subject.

3. P1, L25: What is a "The single-column problem"?

4. P1, L51: What is "a population-balance equation"? Please add references. Isn't it the Boltzmann transportation equation conserving mass?

5. P2, L10: Do you mean "turbulent mixing"? Numerical approach?

6. P2, L15: Please be more specific about the challenging physical processes. What is "represent the subtleties"?

7. P2, L20: Please cite "https://doi.org/10.1146/annurev-fluid-011212-140750". "inherent limitations" of what?

8. P2, L35: in determining both the ...

9. P2, L40: Please add references to the statement "The parameterisations used

in climate models are developed based on smaller-scale simulations resolving particle-size spectrum evolution.". Small-scale models can simulate the cloud microphysical processes while GCMs cannot. Can you give an example on which GCM adopts processes simulated using which small-scale model?

10. P2, L75: What is "title ="? What do you mean here?

11. P2, L105: droplets?

12. P3, L5: by comparing to ...

13. P3, L5: the upwind ...

14. P3, L25: praised. What do you mean by "prediction accuracy of ..."?

15. How do I connect the paragraphs above and below the paragraph "Aerosol Science: ..."? What is the scientific question you want to summarize from this book?

16. P3, L40: I don't understand this sentence. Please rephrase.

17. P3, L45: fixed and moving bin-approaches

18. P3, L50: What do you mean by "...at.., ...at..."?

19: What do you mean by "a grid composed of 2000 size bins"? Is it the spatial resolution? 20: P3, L90: I don't understand the statement of "there is a degree of freedom in the choice of the particle-size parameter used as the coordinate". n(r), n(s), and n(v) are exchangeable. It is a matter of preference of using the radius binning or mass binning. How is it related to "a degree of freedom"? The same comment applies to "p(r(x))".

21: P4, L20: The sentence is incomplete.

22: Caption of Fig.1: What is "ln2(r^3)"? To improve the readability of Fig.1 and 2, you could move the title to the y-axis and use the abbreviation "m, cm, um" instead of words. Please check all the figures and improve the labels and units presented in them.

I still don't understand the purpose of showing both Fig.1 and 2. Fig.2 looks almost identical to Fig.1. If you want to compare the two cases, you should plot them in a same figure.

23: P5, L10: Which figures are you talking about? This paragraph is repeating what is said in the caption of Fig.1. What is the point of repeating it?

24: P5, L15: What do you mean by saying "integrating the number conservation law"?

25: At least the last three paragraphs of section 2.2 can be merged into one. You may describe and explain the results in a logical way. This is just one example, please carefully read through the description of other figures and make the storyline fluent and concise.

26: P6, L5: Which "resulting in" which?

27: P6, L10: What is "sought modified equation"?

28: P6, L55: What do you mean by "transported signal" and "variable sign signals."? Numerical formula. Please rephrase this sentence. The grammar is not correct.

29: P6, L65: About this statement "Overall,

while the MPDATA solutions are superior to upwind, the

drop in amplitude and broadening of the resultant spectrum

still visibly differs from the discretised analytical solution.", is it because of the hyper-diffusion method used in MPDATA or the bin resolution? The upwind scheme presented in Fig.1 of

"https://doi.org/10.1002/2017MS000930" also underrepresents the amplitude of peaks.

30: P7, L5: What do you mean by "linearising MPDATA about an arbitrarily large constant"? It should be "background scalar field".

31: P7, L15: "..., such". It should be a new sentence "Such ...".

32: You can combine Fig 1, 2, 3, and 4 into one figure to improve the readability. Please go through the manuscript carefully and make the figures concise.

33: P7, L25: Are the "negative values" generic of the "infinite gauge" or just for your case? Please explain why they are negative.

34: The last 3 paragraphs in the section 2.11 can be merged into one.

35: P10, L10: "right panel"  $\rightarrow$  "rhs panel". Please check "right" and "left" expressions all across the manuscript.

36: P10, L20: "so" → "such that"

37: P10, L25, L30: I don't understand this paragraph. Please check the grammar.

38: P11, L10: "what" → "that"

39: P11, L55: What is the "Ordinary particle volume concentration"?

40: P12, L30: Is "w1" the initial vertical velocity?

41: P12, L75: How is the vertical velocity determined?

42: The caption of Fig.10 does not read right.

43: The first paragraph of section 3.3 describes the numerical setup. It can be moved up to the one-sentence paragraph.

44: Fig.11: It is hard to distinguish different lines in the vertical-profile plots. Can you use different symbols? Why does "MPDATA iterations: 1" yield different d compared with "MPDATA iterations: 2" and "MPDATA iterations: 3".

45: P13, L40: "the only conclusion here is that the visualisation method used in

Fig. 10 is apt to highlight this feature". This statement again questions the application of the singlecolumn simulation in this manuscript. I still don't understand if the conclusion about the MPDATA from the box simulation can be carried over to the single-column simulation. I encourage the authors to discuss this link in depth, which can be a highlight of this study. Otherwise, I don't see the point of including a very simplified simulation of the single-column model.

46. P15, L15: This study focuses on ...