

Interactive comment on “Technical Note: A new Size REsolved Aerosol Model (SIREAM)” by E. Debry et al.

E. Debry et al.

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General comment of reviewer :

This paper presents a detailed description of a bin-resolved aerosol model which solves that part of the General Dynamics Equation (GDE) concerned with clear-air processes (i.e. aerosol nucleation, coagulation and condensation/evaporation). The paper presents a very thorough description of the physical parameterizations and numerical techniques used in the SIREAM model. A brief description of the cloud and heterogeneous processes which are not directly part of SIREAM is also included. On the whole the paper is well written and the detailed numerical methods employed by the model are mostly clearly described.

My main criticism is that the methods presented are often not sufficiently well refer-

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enced. For instance in section 3.2.1, the coagulation equations use the method described in Jacobson et al (1994) except that the time integration is carried out as 2nd order explicit (described in section 3.3.1) rather than the semi-implicit formulation in Jacobson et al (1994). Also, the section describing the aqueous chemistry in clouds (2.3.1) is at times unclear and needs substantial clarification (see specific comments). However, the detailed description of the parameterizations and numerical methods used in the model is to be welcomed and the Technical Note will be a worthy addition to ACP once the specific comments have been properly addressed.

We thank the reviewer for her/his general comments which helped us to improve the general aspect and the understanding of our paper. We repeat the reviewer's statements below and comment on them:

Specific Comments ———

1) Abstract - page 11846, lines 5-7 - state clearly that the hybrid method refers to a hybrid equilibrium/dynamic approach to mass transfer.

We replace "hybrid method" by "hybrid equilibrium/dynamical mass-transfer method" in the revised version of our article.

2) Introduction – page 11847 line 8 - replace "rigorous" with "detailed". Simpler models can still be rigorous.

Right ! We replace "rigorous" by "detailed" in the revised version of our article.

3) Introduction – page 11847 lines 15-20. The authors should include references to the other sectional models in the literature (see e.g. Jacobson, 1997; Adams and Seinfeld, 2002; Gong et al, 2003; Spracklen et al, 2005) and explain how the parameterized processes and/or numerical treatment described in the paper differs from these.

The above references have been added to the revised version, however our purpose in this article is not to present a review of aerosol models, but just to present our aerosol model.

4) Introduction – page 11847 line 22 – do the authors really mean SIREAM is "strongly coupled" to MAM or are the two models rather alternative aerosol models within "ATMODATA".

MAM and SIREAM are, as the referee says, two model alternatives sharing the same physical parameterizations, gathered in ATMODATA. We replace "strongly coupled to a companion model" by "SIREAM is the size-resolved alternative to the modal model".

5) Introduction – page 11847 line 26 – the paper should state whether the model is intended for use in Lagrangian or Eulerian (global or regional?) frameworks.

This model has been used in Eulerian framework at global, regional and local scale. We add "... used in several global, regional and local eulerian applications".

6) Section 2.1 - page 11849 line 10 - after the description of the method for determining the inorganic composition, the method for determining the organic composition should be included (i.e. add a sentence stating that the SORGAM scheme is used [with reference to the Schell papers]).

We explicitly cite Schell paper in this section but do not give further details at this point of the text as the organic composition and SORGAM model are fully described in section 2.2.5.

7) Section 2.2 - page 11850 line 16 - the expression used to calculate the coagulation kernel should be stated here – it is not explained anywhere in the paper.

To our opinion, adding the expression of the coagulation kernel would load down the article without making it clearer. Indeed the expression of the coagulation kernel is not needed in the article, on the contrary to the condensation kernel one that we use in the numerical part. The brownian coagulation kernel expression is also rather well-known and can be found in most of aerosol books.

8) Section 2.2.3 - page 11851 line 11, Section 2.2.4 page 11852 line 9 – the Nenes et al (1998) reference for ISORROPIA should be included here for easy reference.

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We add the reference for ISORROPIA at both places in the revised version.

9) Section 2.3 - page 11857 line 14, - the authors state that the processes are not related to the General Dynamics Equation. But this is not really the case since both will affect the size distribution. Better to state that the processes are not part of SIREAM.

We replace "not directly related" by "not directly part".

10) Section 2.3 - page 11857 line 17, - this section should explain how these other processes are treated in the model with respect to the main SIREAM code - are they solved process-split? The explanation could go either here or perhaps better in section 3.1.

There must have a mis-understanding here as the aqueous model is an alternative to the SIREAM code in case of the presence of clouds. That is to say that within a cell either the aerosol module takes place or the aqueous model, but not both. Maybe we do not state this clearly, so we add at beginning of section 2.3.1 : "For cells with a liquid water content exceeding a critical value ..., the grid cell is assumed to contain a cloud and the aqueous-phase module is called instead of the SIREAM module." In what concerns heterogeneous reactions we had already written : "For numerical stability requirements, these reactions are coupled to the gas-phase mechanism."

11) Section 2.3.1 - page 11858 line 5 and 13. – I strongly agree with the comments of the other referee regarding the use of a critical activation diameter of 0.7 microns. The description suggests that bins larger than this (dry?) diameter be mapped on to a monomodal (lognormal?) distribution with median diameter 0.4 microns. This seems inconsistent to me – please can the authors clarify.

This part seems to have raised the attention of both referees for some elements were indeed lacking. Let me explain more clearly. The upper part of the aerosol size range (above the activation diameter) is transferred to the aqueous phase when a cloud exists. The aqueous phase consists in our model of a bulk phase (no sections although it is

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possible). The cloud droplet diameter is assumed to be equal to 20 micrometers during aqueous chemistry computation. There is no modal representation until this step. After computation, it is necessary to redistribute the bulk aqueous mass onto the aerosol size sections. To do so it is assumed that the aerosol distribution has a bi-modal shape (not mono modal as we stated at first). This assumption is only used to compute the fraction of the bulk aqueous mass that will fall in each activated aerosol sections. The median diameter and variance for both modes are respectively 0.4 micrometers and 1.8 for first mode, 2.5 micrometers and 2.15 for second one.

We rewrite this section as clearly as possible in the revised version.

12) Section 2.3.1 – page 11858 lines 13/14 – it seems strange that there is little sensitivity to the choice of the size distribution assumed for the activated particles. The two references given for the aqueous chemistry (Fahey, 2003; and Fahey and Pandis, 2001) are also not easily available, so the reader is not able to understand why this is so. Please can the authors explain.

Maybe the referee thinks about the size distribution of cloud droplets which does have an impact on aqueous chemistry computation as pointed out by Kathleen Fahey's recent articles. Nevertheless our remark deals with the size distribution used for the redistribution step (bimodal shape). This one has been shown to be little sensitive, according to Kathleen Fahey's phd-thesis.

13) Section 2.3.1 - page 11858 line 15 - the authors should state (as in Sportisse et al, 2006) that for the purpose of cloud-chemistry, the cloud droplets are assumed to have a fixed size of 20 micron dry diameter.

We add "the average diameter of droplets is fixed to 20 micrometers" in the revised version.

14) Section 2.3.1 – page 11858 lines 17/18 - it is not clear to the reader which parts of the mass transfer to the cloud droplets is solved dynamically and which is solved as-

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suming equilibrium. I suggest that the authors include Table 1.2 [page 22] in Sportisse et al (2006).

I agree that this section would be clearer with this table. However it would load down the article with a quiet heavy table which is not directly part of SIREAM. We add an explicit reference to Sportisse et al (2006) at this point of the text, this reference is also easy to retrieve from polyphemus web site.

15) Section 2.3.1 - page 11858 lines 26/27 – this sentence is not clear – Sportisse et al (2006) describe the diphasic model that is used. This should be briefly described if used along with a clear description of the mapping of the cloud droplet distribution onto the particle distribution.

This remark seems to me to be connected to remark 11. I hope this section will appear clearer with answer to remark 11. We rewrite this paragraph in the revised version as following : "After one timestep, the new mass generated from aqueous chemistry is redistributed onto aerosol bins that were activated. To do so the initial aerosol distribution is assumed to have a bimodal shape (log-normal distributions) that gives weighting factors for each aerosol bin. Median diameter and variance for each mode are respectively 0.4 micrometers and 1.8 for first one, 2.5 micrometers and 2.15 for second one. The tests in Fahey (2003) illustrate the low impact of the choice made for this assumption."

16) Section 3.1 - page 11860 lines 8/9 – I strongly support the other referee's comment that when the redistribution onto a fixed size grid is used (as required in a 3D model), SIREAM is effectively using the quasi -stationary method of Jacobson (1997) rather than a moving-sectional method. Furthermore I do not think it is entirely accurate for the authors to state that the method avoids numerical diffusion. The quasi-stationary method will avoid *some* of the numerical diffusion associated with the full-stationary method but will not perform as well as the moving-centres or full-moving methods, as shown by Jacobson (1997). The authors should revise their statements accordingly (and other places this is stated in the text - (heading page 11862 line 2).

Point taken. Nevertheless the "Lagrangian method" refers for us only to the fact that bins are allowed to grow due to condensation/evaporation, whatever is the redistribution process. On the contrary "the quasistationary method" refers to both a lagrangian approach and a particular way to redistribute lagrangian bins on a fixed grid. Furthermore we propose a different way to redistribute based on a new way to compute bounds.

Thus we rewrite the paragraph as follows: "Third, condensation/evaporation is solved with a Lagrangian method, the quasistationary method of Jacobson (Jacobson 1997) is employed to reduce the numerical diffusion associated with Eulerian schemes in the case of a small number of discretization points (typically the case in 3D models)."

17) Section 3.2.2 – page 11862 line 2 - section 3.2.2. needs an opening paragraph to introduce this section as it provides the main new method in the paper. The two alternative methods should be clearly introduced and I agree with the other referee that once again, since the 2nd method is essentially the quasi-stationary method as in Jacobson (1997) it should be referenced and stated as such. The text should also describe in which situations each of the methods should be used (with advantages/disadvantages).

I agree that second method comes to the fitting method developed by Jacobson (Jacobson 1997). We add "This algorithm comes to the fitting method developed by Jacobson in (Jacobson (1997))." We also points out to our opinion the advantages/disadvantages of both methods.

18) References: page 11873 - the Koo et al (Aer Sci Tech) paper should have authors Koo, Gaydos and Pandis – please amend accordingly.

19) References: page 11874 - the Sartelet et al (Aer Sci Tech) reference was published in 2006 - plesae revise accordingly.

References not in manuscript —————

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Spracklen, D. et al, 2005, A global off-line model of size-resolved aerosol microphysics: 1: Model development and prediction of aerosol properties, *Atmos. Chem. Phys.*, 5, pp. 1-26.

We corrected and added all above references.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 6, 11845, 2006.

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