# Numerical investigations on the modelling of ultrafine particles in SSH-aerosol-v1.3a: size resolution and redistribution

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**Reply to Anonymous Referee #3's comments**

*I am joining the review process after the paper has undergone one round of reviews. While the revised version addresses some of the reviewers' concerns—most notably the addition of the 0D box model verification strengthens the paper—several significant weaknesses persist. These weaknesses may be challenging to resolve sufficiently to make the paper publishable. Below, I provide detailed comments regarding the methodology, nomenclature, and presentation:*

*1. Research Gap: I agree with the other reviewers that it is unclear if the paper effectively addresses a critical research gap. From the box model simulations, we learn that "the Lagrangian scheme is able to achieve a similar accuracy to the one obtained with the Eulerian scheme using a twofold resolution" (line 170), but also that "the trade-off to pay in terms of computational time, when choosing the Lagrangian scheme rather than the Eulerian one, is a factor of about two to three" (line 171). However, the analysis lacks a cost-error plot—a standard tool for evaluating the performance of new algorithms. If my understanding of the results is correct, running the Eulerian scheme at twice the resolution would achieve a similar error at a comparable computational cost. If this is the case, it raises the question of the new algorithm's purpose and benefits.*

Our reply: The research gap has been better highlighted in the introduction, by adding the following sentences:

> The use of a large number of sections in CTMs is challenging because each section can contain multiple chemical species. As a result, the number of transported compounds in the Eulerian model is equal to the number of chemical species multiplied by the number of sections.
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> ...
>
> Hence, "moving sectional" models are designed to resolve condensation and evaporation processes (Kim and Seinfeld, 1990). However, modeling coagulation is essential to represent the formation of ultrafine particles.
>
> ...
>
> Here, an analytical expression is derived under the assumption of uniformly distributed particles within each section. This allows the development of a moving sectional model that can resolve all processes related to aerosol dynamics.

The new algorithm's purpose and benefits have also been strengthened by additional analysis of the box model simulations. To better highlight the advantages of the new scheme, two types of errors are now considered: the relative error on the integrated number concentration (as in the previous version) and the relative error on the number distribution. The second error indicator is able to penalize more significantly dynamics which are smoothed out compared to the reference, which is indicative of larger numerical diffusion. In the box-model simulation, for particles of diameters lower than 10 nm which are faster evolving, the Lagrangian scheme achieves lower errors for a given number of sections. The Eulerian scheme achieves a similar accuracy to the one obtained with the Lagrangian scheme only for particles of diameters higher than 10 nm. When accounting for error as a function of execution time, the Lagrangian scheme is indeed penalized by its larger computation needs. Additional analysis with cost-error plots were added to section 3. The plots clearly demonstrate that the relative error of the Lagrangian scheme is significantly lower than that of the Eulerian scheme for particles with diameters below 10 nm—where aerosol dynamics are most active. Since the new scheme provides a more accurate prediction of the size distribution, the differences between the two schemes are more pronounced for the relative error on the number distribution. The following lines have been added to section 3.1:

> The distributions obtained with both schemes are compared in terms of relative error against the reference simulation using 200 sections. Figure 2 shows the relative errors on integrated aerosol number concentration, while Figure 3 shows the relative errors on aerosol number distribution. For particles in the range 1 - 10 nm, the dynamic mesh scheme consistently outperforms the fixed mesh scheme, yielding lower errors for both error indicators. The difference between the two schemes is more pronounced when comparing relative errors in number distribution, rather than errors in integrated number concentrations. This suggests that the enhanced performance is due to the less smoothed aerosol distribution. For particles with diameters higher than 10 nm, both the fixed and dynamic mesh coagulation schemes produce similar errors for a given number of sections, with errors decreasing as the number of sections increases. The similarity between both schemes in this diameter range is expected, as the time evolution is much slower. However, the dynamic mesh coagulation scheme requires more computational time than the fixed mesh coagulation scheme for a given number of sections, as it necessitates frequent re-discretizations of the coagulation operator. Figures 4 and 5 show the errors as a function of execution time for different number of sections. The overall trends are similar for both schemes, with an increase in execution time and a decrease in error as the number of sections increases. For particles of diameters in the 1-10 nm range, although the dynamic scheme requires more computational time than the fixed scheme, it achieves lower error values, particularly in the number distribution. In contrast, the fixed scheme shows only a slow reduction in errors. For particles of diameters larger than 10 nm, both schemes yield very similar results in terms of accuracy, as there is little evolution in this size range. Consequently, the dynamic mesh is disadvantaged by its higher computation time. As a result, the curves representing the dynamic mesh scheme in Figures 4 and 5 appear as horizontal translations of those representing the fixed scheme. This highlights that the advantages of a more complex scheme are only justified in regions where aerosol dynamics are most active.

*2. 3D Model Implementation: The paper suggests that some remapping occurs when implementing the algorithm into the 3D model (line 180). However, the impact of this remapping on the simulation of size distributions is unclear. I recommend designing a 0D test case that replicates the exact operations performed in the 3D model (e.g., redistribution every 100 s) and using this case to produce a cost-error plot. This approach would help clarify the implications of the remapping process on model performance and accuracy.*

Our reply: We would like to thank you for this suggestion. We have added a new intermediate scheme in the 0D-box comparisons, which corresponds to the Lagrangian scheme with redistribution every 100 s, to replicate the operations performed in the 3D model. Furthermore, we have also studied the evolution of accuracy as a function of the redistribution timestep. We show that the intermediate scheme deviates from the Eulerian scheme only when the redistribution timestep or the resolution is large enough. The scheme behaves similarly to the Lagrangian in the limit of large redistribution timestep and number of sections.

The following lines have been added to section 3.2:

> An intermediate scheme is added to the 0D-box comparisons. It corresponds to the dynamic mesh scheme with redistribution every 100 s, to replicate the operations performed in the 3D model.
>
> As shown in Figures 2, 3, 4 and 5, the results of the dynamic mesh are very closed to those of the fixed mesh in terms of errors, if redistribution is applied every 100 s. In that setting, the dynamic mesh scheme loses some of its advantage, as the introduced diffusive step brings its performance closer to that of the fixed mesh scheme compared to the unperturbed dynamic mesh scheme. Figure 6 illustrates how the mean relative error evolves with different redistribution timesteps. In the limit of a large number of sections and a large redistribution timestep, the intermediate scheme behaves similarly to the dynamic mesh scheme. However, as the redistribution timestep decreases, diffusivity increases, negatively impacting the scheme's performance, making it comparable to the fixed mesh scheme but with a higher computational cost. This implies that in a 3D setting, the dynamic mesh scheme may offer similar effectiveness to the fixed mesh scheme when fluid dynamics are modeled within an Eulerian framework, depending on the number of sections and redistribution frequency. However, the dynamic mesh scheme would provide greater advantages in Lagrangian transport models.

And the following lines have been added to the conclusion

> However, 0D simulations have shown that the regular redistributions imposed by the assumptions of the 3D Eulerian model significantly limit the efficiency of the dynamic mesh algorithm. While in a 0D setting, this algorithm greatly reduces errors for particles strongly affected by aerosol dynamics, its advantages are diminished in the 3D Eulerian framework. Hence, it would be more suitable to use the algorithm in Lagrangian transport simulations, which deal with advection in physical space in a Lagrangian fashion.

*3. Nomenclature: I agree with Reviewer 2 that the term "Lagrangian" might be misleading, as it could be confused with particle-based or super-particle methods commonly described in the literature. The term "moving sectional" model, as used*

*by Kim & Seinfeld (1990), may be a more accurate and appropriate descriptor for the type of model employed in this study.*

Our reply: We now refer to the proposed algorithm as 'dynamic mesh coagulation' and to the pre-existing method as 'fixed mesh coagulation'. This name should avoid confusion with particle-based methods by putting emphasis on the fact that our method relies on a Lagrangian description of the underlying aerosol volume mesh.

> In this paper, the 'dynamic mesh coagulation' algorithm is proposed and implemented in the aerosol dynamics model SSH-aerosol. It features a Lagrangian dynamic discretization of the aerosol size range, which evolves in accordance to the evolution prescribed by condensation and evaporation. Coagulation is solved on the resulting dynamic mesh by use of a time-dependent discretization of Smoluchowski equation.

We would like to thank you for the comment, which helped us to better formulate the research gap question. The 'moving sectional' method developed by Kim & Seinfeld is significantly different from the method we propose, since authors clearly state that their approach is suitable when accounting only for condensation/evaporation. Our approach allows to represent both condensation/evaporation and coagulation processes under a common description, and addresses a gap in previously developed numerical methods for solving the full aerosol dynamics equation. The introduction was modified accordingly, as detailed in the reply to the first comment.

*4. Writing Style: While some typos appear to have been corrected, the overall quality of the writing remains substandard and detracts from the paper's readability. Many sentences are awkwardly phrased, such as "As the health impact of ultrafine particles is getting better understood…" and "The condensation process is formally equivalent to advection in aerosol volume." A more thorough review of the language and style is necessary to meet publication standards.*

Our reply:

We have improved the wording of the paper. For example, in the section where we introduce the implications of choosing an Eulerian or a Lagrangian framework for condensation/evaporation and coagulation (line 42).

> The condensation/evaporation process is formally equivalent to advection in aerosol volume. One of the main drawback of the classical Eulerian framework to solve advection equations is the introduction of numerical diffusion. The Lagrangian approach which aims at limiting numerical diffusion that would be introduced by the numerical discretization in an Eulerian frame of reference is therefore often applied (Neuman, 1984; Seigneur et al., 1986; Tsang and Rao, 1988; Gelbard, 1990). This Lagrangian approach is however conflicting with the Eulerian one often used to solve the coagulation process, which involves interactions between different aerosol size ranges (Gelbard et al., 1980).
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> Condensation and evaporation behave like a transport process, moving particles within the aerosol volume space, as they grow or shrink while interacting with the gaseous phase. One of the main drawback of the classical Eulerian framework when solving advection equations is the introduction of numerical diffusion. The Lagrangian approach is often applied in that context (Neuman, 1984; Seigneur et al., 1986; Tsang and Rao, 1988; Gelbard, 1990) in an

effort to alleviate the effects of numerical diffusion, which would be introduced by the numerical discretization in an Eulerian frame of reference. Using Lagrangian approach to represent the aerosol size discretization conflicts with the Eulerian framework typically chosen to solve aerosol coagulation, which relies upon a fixed discretization through time. To solve both coagulation and condensation/evaporation, models are required to switch between Lagrangian and Eulerian frameworks, introducing numerical diffusion which may hinder numerical performance.

In the abstract, the sentence starting by "As the health impact of ultrafine particles is getting better understood. . . " is replaced by

As the health impacts of ultrafine particles become better understood, accurately modeling size distribution and number concentration in chemistry transport models is becoming increasingly important.

## References


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