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 #2's comments

Jacquot and Sartelet describes a semi-Lagrangian approach for representing the evolution of the aerosol size distribution through coagulation. They implemented this approach in the SSH-aerosol model and quantified the impact on simulated aerosol number concentrations over Greater Paris by coupling the updated ssh-aerosol scheme with a 3D chemical transport model. In these simulations, they quantified the sensitivity of the simulations to the resolution of the size distribution. While the technical issues with this paper (described below) may be fixable, I am not convinced that the study addresses a critical research gap, and the technical approach is rather weak. The paper is also poorly written with many grammatical errors. Further, I do not understand why this paper is being considered for the special issue on particle-based methods; they describe an algorithm for a sectional model. I do not see how this is a particle-based method.

Our reply: By using the new scheme, which is free of numerical diffusion, this work allows to provide a relative assessment of different error sources for ultrafine particle modeling. We observe that numerical diffusion is dominated by the errors related to the coarseness of the discretization used to represent the coagulation operator. To better specify the objective of this study, line 77 of the introduction

"In order to evaluate the significance of errors introduced by numerical diffusion during the coupled integration of coagulation, condensation-evaporation and nucleation, a new algorithm coined 'Lagrangian aerosol dynamics' is proposed and implemented, making it possible to do away with the Eulerian approach to solve coagulation. To remain in a Lagrangian frame of reference, the representation of coagulation needs to be dynamically adapted to the size mesh evolution. The proposed algorithm, which avoids redistribution when solving aerosol dynamics, is presented in section 2. The chemistry-transport model used to assess the impact on concentrations and the setup of the 3D simulations are presented in section 3. Finally, the impact of different size resolutions and of the new algorithm are presented in section 4."

was replaced by:

"A new algorithm, termed 'Lagrangian aerosol dynamics,' is proposed and implemented to enable a comparative assessment of key error sources in ultrafine particle modeling—specifically, size discretization and numerical

25   diffusion. By replacing the Eulerian approach for solving coagulation, this method isolates and evaluates the impact of numerical diffusion. To limit numerical diffusion and to remain in a Lagrangian frame of reference, the representation of coagulation needs to be dynamically adapted to the size mesh evolution. The proposed algorithm, which avoids redistribution when solving aerosol dynamics, is presented in section 2, along with a 0D-validation. The chemistry-transport model used to assess the impact on concentrations and the setup of the 3D simulations

30   are presented in section 3. Finally, the impact of different size resolutions and of the new algorithm are presented in section 4."

The paper introduces an algorithm designed to improve simulations of the aerosol size distribution, but they do not include any benchmarking for this algorithm. They show differences between their approach and a traditional sectional modeling approach, and they also quantify the impact of these differences for varying resolution of particle sizes. However, they do

35 *not show how their approach compares to analytical solutions, quantify performance against a benchmark model, or even show convergence of their solutions. They use the 25-bin sectional approach as their benchmark, but they do not show any verification of the 25-bin model. They evaluate their findings with observations, but they do not include any verification of their new algorithm against a benchmark model, which would be more appropriate.*

<u>Our reply</u>: We have added an additional section which provides numerical validation of the Lagrangian scheme on a benchmark

40 in the case of an idealized 0D box model. We show the effectiveness of the scheme, which is able to provide results consistent with an Eulerian reference run at a larger resolution.

Numerical validation

To validate numerically the Lagrangian scheme, and to illustrate the difference between the Lagrangian and the Eulerian schemes for different size resolutions, an idealized box setting is considered. The initial mass and number

45   distribution of particles corresponds to the sum of three lognormal distributions of the hazy case of (Seigneur et al., 1986). Particles are assumed to be made of sulfate. To favor nucleation and condensation, gaseous sulfuric acid and extremely low volatile organic compounds formed from the autoxidation of monoterpene (Chrit et al., 2017; Sartelet et al., 2020) are initialized with concentrations of $2 \cdot 10^{-2}$ μg.m^{-3}. Temperature is set to 27°, pressure to 1 atm and relative humidity to 40%. A one hour simulation is performed, using different size resolution levels. All

50   gaseous species have either condensed or nucleated at the end of the simulation. This test case is highly stringent for number concentrations, as the gaseous concentrations result in intense nucleation. For each configuration, particles range from 1 nm to 10 μm and the size distribution is geometrically refined using either 4 sections, 12 sections, 25 sections or 50 sections. The reference simulation is a simulation performed with 200 sections using the Eulerian scheme.

55   Figure 2 and Table 2 highlight the very good agreement of both schemes on mass concentration for all size resolution. This is due to the idealized configuration of the test case, with non-volatile compounds only. The accuracy of the Eulerian scheme is nearly independent of the size resolution and relative errors reach the order

of $10 \cdot 10^{-10}$, the accuracy of the Lagrangian scheme quickly decreases from a strong relative error baseline of $10 \cdot 10^{-7}$ at only 4 sections.
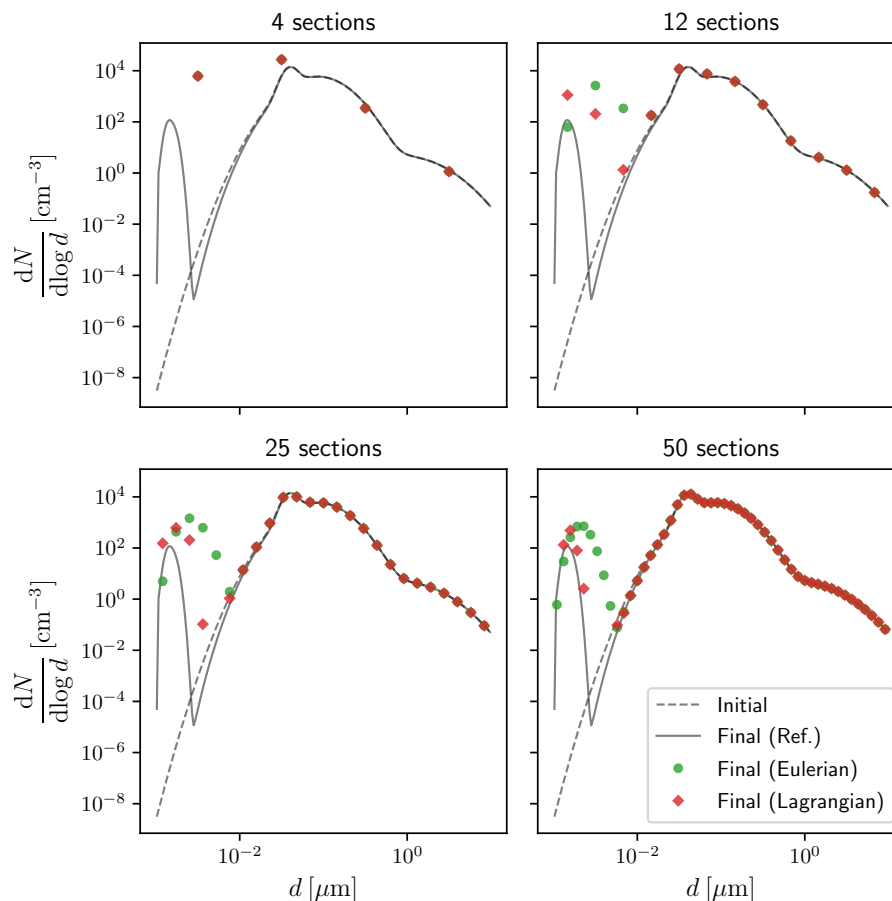

Figure 1. Evolution of the number concentration simulated with the Eulerian and Lagrangian schemes at different size resolutions. The reference is computed with the Eulerian scheme using 200 sections.

For number concentrations, larger differences are observed between the schemes and the size resolution than for mass concentrations. Figure 1 and Table 1 illustrate the differences between the number concentrations simulated with the two schemes at different size resolutions. The differences with the reference simulation increase as the size resolution decreases. The differences are particularly high for 4 sections (about 460% for the number concentration), but they are much lower for 50, 25 and 12 sections (0.8%, 6% and 38% for the number concentration with the Lagrangian scheme for 50 and 25 sections respectively).

The Lagrangian scheme leads to improved accuracy, particularly for particles with a diameter lower than 10 nm. For the number concentration of particles with a diameter smaller than 10 nm, and for 12 sections and higher, the

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Lagrangian scheme is able to achieve a similar accuracy to the one obtained with the Eulerian scheme using a twofold resolution. The Lagrangian scheme still outperforms the Eulerian scheme on total number concentrations, but not as strongly as for number concentrations of particles with diameter smaller than 10 nm. 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.
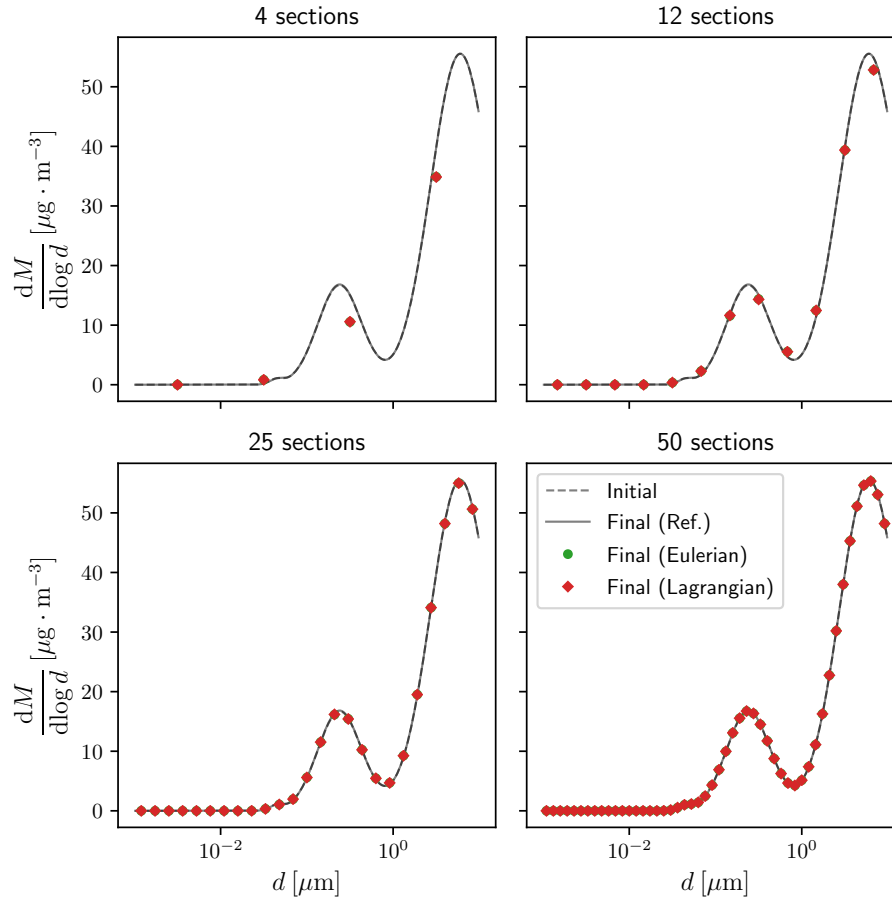

Figure 2. Evolution of the mass concentration simulated with the Eulerian and Lagrangian schemes at different size resolutions. The reference is computed with the Eulerian scheme using 200 sections.

Further validation is provided by the comparison to observations in a 3D context, already present in section 3.2 of the manuscript.

The paper describes their algorithm as "Lagrangian", but they project the size distribution onto a fixed grid at every time step. As there are several truly Lagrangian aerosol and models in the literature (e.g., Shima et al., 2009 and Riemer et al.,

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	Relative error on number concentration (absolute value)					
	$d \leq 10$ nm		$d > 10$ nm		Total number	
Resolution	Eulerian	Lagrangian	Eulerian	Lagrangian	Eulerian	Lagrangian
4 sections	418	405	3.58	3.58	4.61	4.58
12 sections	66.5	28.5	$3.12 \cdot 10^{-1}$	$3.10 \cdot 10^{-1}$	$4.76 \cdot 10^{-1}$	$3.80 \cdot 10^{-1}$
25 sections	26.3	9.34	$3.84 \cdot 10^{-2}$	$3.82 \cdot 10^{-2}$	$1.04 \cdot 10^{-1}$	$6.14 \cdot 10^{-2}$
50 sections	10.3	2.78	$1.87 \cdot 10^{-3}$	$1.53 \cdot 10^{-3}$	$2.74 \cdot 10^{-2}$	$8.42 \cdot 10^{-3}$

Table 1. Relative error (absolute value) for different size resolution for number concentration, estimated with the Eulerian and Lagrangian schemes. The reference is computed with the Eulerian scheme using 200 sections.

	Relative error on mass concentration (absolute value)	
Resolution	Eulerian	Lagrangian
4 sections	$2.14 \cdot 10^{-10}$	$1.3 \cdot 10^{-7}$
12 sections	$2.42 \cdot 10^{-10}$	$1.45 \cdot 10^{-8}$
25 sections	$1.89 \cdot 10^{-10}$	$6.66 \cdot 10^{-9}$
50 sections	$1.46 \cdot 10^{-10}$	$5.10 \cdot 10^{-9}$

Table 2. Relative error (absolute value) for different size resolution for mass concentration, estimated with the Eulerian and Lagrangian schemes. The reference is computed with the Eulerian scheme using 200 sections.

2009), I found this characterization misleading. I think it would be more accurately described as "semi-Lagrangian". I do not understand why this approach is being described in a special issue on particle-based methods.

80 <u>Our reply:</u> Our approach is Lagrangian in aerosol volume, and would indeed be fully Lagrangian in a 0D setting where no spatial dependency is accounted for. For models which account for spatial inhomogeneity such as 3D chemistry-transport models, other processes such as advection and diffusion of air masses inevitably require that all cells share the same aerosol volume discretization. To distribute the proposed algorithm in a realistic 3D setting, a projection step to the original volume mesh is therefore necessary. However, this projection only occurs at timesteps during which neighboring cells communicate.

85 In a 3D setting, internal dynamics in each cell is treated in a fully Lagrangian manner, between timesteps enforced by the 3D CTM.

However we do agree that our method is not particle-based, and we do not claim so. Furthermore, the referee refers to approaches which are Lagrangian in space (following the spatial evolution of each particle), but they are not Lagrangian in aerosol volume. The framework of the Smoluchowski equation describing coagulation depends on aerosol volume and time,

90 and we studied the impact of the Lagrangian formulation in aerosol volume.

The authors state that the numerical results are more sensitive to the resolution of the size distribution than the incorporation of their new algorithm. Even with substantial revisions, the impact of this paper as it is currently framed seems limited.

Perhaps it would be better to reformulate the paper to focus on the impact of the size resolution, rather than advocating for a new algorithm that has a relatively small impact on the simulation results.

Our reply: We aim to evaluate the relative magnitude of different error sources related to different processes. Simply increasing the resolution does not bring significative knowledge as to which potential error source dominates the remaining numerical errors. By resorting to a fully Lagrangian formulation at different resolution levels, we are able to asses the relative importance of errors due to the coarseness of the discretization of the coagulation operator compared to the numerical diffusion introduced by the Eulerian formulation. For typical urban dynamics and at resolutions usual in a 3D setting, we show that numerical diffusion is not the dominant error source.

Since the authors state that the impact of their semi-Lagrangian scheme has the greatest impact in their low-resolution simulation of 9 bins, I wonder if the proposed algorithm may be more relevant for extremely low-resolution sectional schemes (e.g., 4 bins). WRF-Chem, for example, is often run with 4 sections.

Our reply: In our 0D investigation, we showed that 4 sections models are generally too coarse to represent accurately aerosol dynamics for ultrafine particles. Such a low resolution is not suitable for accurate simulations where the number concentration of ultrafine particles is of interest. Our scheme enables to mitigate effects related to numerical diffusion associated to condensation-evaporation, but cannot mitigate issues related to an inaccurate representation of other processes such as coagulation, which is inevitably a concern with such low resolution.

The references in the introduction should be double-checked. For one thing, the aerosol chemistry model is "MOSAIC" not "MOSAIC", and the correct citation is "Zaveri, R. A., Easter, R. C., Fast, J. D., & Peters, L. K. (2008). Model for simulating aerosol interactions and chemistry (MOSAIC). Journal of Geophysical Research: Atmospheres, 113(D13)." I noticed this error in the references because the model was misspelled, but I suggest double-checking to be sure the correct paper is referenced.

Our reply: We have traded all occurrences of "MOZAIC" for the correct acronym "MOSAIC", and have modified the reference as suggested. Line 70

The SSH-aerosol model (Sartelet et al., 2020) is used to solve the general dynamics equations describing aerosol evolution. Coagulation, nucleation, condensation of extremely-low volatile organic and non-volatile compounds are solved simultaneously. The condensation/evaporation of semi-volatile aerosols is modeled using either a dynamic or a bulk equilibrium approach, assuming instantaneous thermodynamic equilibrium between the gas and bulk-aerosol phases. In the bulk approach, the size-section weighting factors depend on the ratio of the mass transfer rate in the aerosol distribution; and the Kelvin effect, which limits the condensation of those compounds on ultrafine particles, is modeled following Zhu et al. (2016). Time integration is performed using the trapezoidal rule, an explicit Runge-Kutta method of order 2, with an embedded order 1 method enabling error estimates and adaptive time stepping. For both the Eulerian and Lagrangian schemes, the first step consists in computing the coagulation partition coefficients which are necessary to discretize the coagulation operator.

For the Eulerian algorithm, the evolution of particles due to coagulation is simulated using the pre-computed partition coefficients on the fixed reference grid, while condensation-evaporation are treated in a Lagrangian manner.

After each time step, as the diameters of particles may have evolved because of the Lagrangian formulation of condensation, a redistribution scheme is applied, such as the moving diameter (Jacobson, 1997) or the Euler-coupled scheme (Devilliers et al., 2013). The outline of this implementation is described in Algorithm 1.

To estimate the impact of redistributing every time step onto the fixed Eulerian grid, a Lagrangian algorithm is setup for aerosol dynamics, as described in 2. Coagulation partition coefficients are then computed at the beginning of each timestep, allowing for the size mesh to evolve. Aerosol concentrations evolve in a Lagrangian manner under both coagulation and condensation-evaporation. Contrary to the Eulerian scheme, redistribution is not applied at the end of each timestep. Hence the sections boundaries evolve with time. A safety feature is implemented, such that if section boundaries were to cross, redistribution is applied so that the integration can be followed though on a well ordered partition of the size discretization, which is a necessary condition for partition coefficients to be well defined. Note that, to fit the framework of a 3D CTM, redistribution is always performed at the end of each 0D simulation when t_{final} if reached. This final time corresponds to the timestep of th 3D-model, i.e. the time step used to solve advection and diffusion processes in space. It generally corresponds to multiple timesteps of the internal dynamics of aerosols.

Algorithm 1 Lagrangian integration of condensation and Eulerian integration of coagulation

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Compute coagulation partition coefficients

while $t < t_{\text{final}}$ **do**

    Compute number and mass concentration evolution due to coagulation, condensation/evaporation and nucleation

    Redistribute number and mass concentrations on the fixed Eulerian grid

end while

Algorithm 2 Lagrangian integration of condensation and coagulation

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while $t < t_{\text{final}}$ **do**

    Compute coagulation partition coefficients based on current size mesh

    Compute number and mass concentration evolution due to coagulation, condensation/evaporation and nucleation

    if Some mesh size nodes have crossed **then**

        Redistribute number and mass concentrations on the fixed Eulerian grid

    end if

end while

Redistribute number and mass concentrations on the fixed Eulerian grid

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I suggest adding the units into the headings of Table 1 to improve readability.

145 <u>Our reply:</u> For readability, we have added units to the headings of Tables 1 and 2.

Table 3. Comparison of simulated and measured daily number concentrations of particles $N_{>10}$ between 29 June and 10 July 2009, at the observation sites LHVP and SIRTA, using the Lagrangian scheme. Mean observed ($\bar{o}$) and mean simulated ($\bar{s}$) daily number concentrations are reported in $\#.cm^{-3}$. Fraction of modeled data within a factor of 2 of observations (FAC2) as well as normalized mean bias (NMB) and normalized mean error (NME) are reported in $\%$.

Statistical indicator	SIRTA					LHVP				
	$\bar{o}$	$\bar{s}$	FAC2	NMB	NME	$\bar{o}$	$\bar{s}$	FAC2	NMB	NME
Unit	$(\#.cm^{-3})$	$(\#.cm^{-3})$	(%)	(%)	(%)	$(\#.cm^{-3})$	$(\#.cm^{-3})$	(%)	(%)	(%)
9 sections	5215	4766	75	-9	36	8804	7104	92	-19	30
14 sections	5215	5444	92	4	36	8804	8231	99	-7	29
25 sections	5215	5322	92	2	35	8804	8285	99	-6	28

Table 4. Comparison of simulated and measured daily $PM_{2.5}$ concentrations between 29 June and 10 July 2009, at four available measurement stations available from the AIRPARIF network, using the Lagrangian scheme. Mean observed ($\bar{o}$) and mean simulated ($\bar{s}$) daily mass concentrations are reported in $\mu g.m^{-3}$. Fraction of modeled data within a factor of 2 of observations (FAC2) as well as normalized mean bias (NMB) and normalized mean error (NME) are reported in $\%$.

Statistical indicator	$\bar{o}$	$\bar{s}$	FAC2	NMB	NME
Unit	$(\mu g.m^{-3})$	$(\mu g.m^{-3})$	(%)	(%)	(%)
9 sections	10.4	8.7	94	-10	32
14 sections	10.4	8.9	94	-8	31
25 sections	10.4	9.0	94	-8	30

This paper contains many grammatical errors and typos. Aside from overt errors, the phrasing is often strange and unclear. The technical editing that would be required to bring this paper to a publishable form is beyond the responsibility of a peer reviewer. I strongly recommend sending this paper to a technical editor before resubmission.

<u>Our reply:</u> We have carefully proofread the manuscript to remove some orthographical/grammatical errors and generally im-
150 prove the wording.

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