

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

Oscar Jacquot¹ and Karine Sartelet¹

¹CEREA, Ecole des Ponts, Institut Polytechnique de Paris, EdF R&D, IPSL, Marne la Vallée, France

Correspondence: Oscar Jacquot (oscar.jacquot@enpc.fr) and Karine Sartelet (karine.sartelet@enpc.fr)

Reply to Anonymous Referee #1's comments

5 *General comments: This paper presents the development of analytical equations and a numerical algorithm for Lagrangian calculation of the coagulation term in aerosol distribution dynamics, specifically for calculating the partitioning of coagulated aerosol (number and mass) between discrete size bins in a manner consistent with the Lagrangian formulation of other aerosol dynamics processes. The approach was developed with the purpose of reducing numerical dispersion during aerosol dynamics simulation caused by redistribution due to changing between Lagrangian and Eulerian formulations. In this study, the new approach was applied within a Eulerian chemical transport model to a real-world case study, with three different resolution particle size distribution schemes. Impacts on accuracy (compared with measurements) and numerical dispersion were investigated. The main results suggest that the impact of the new formulation/algorithm on numerical diffusion is small compared*
10 *with the impact of size resolution itself (and compared with the overall error in representing measurements). Overall, I find the work to be interesting and publication of the new more-internally-consistent algorithm is likely to be useful to CTM model developers. However, the impact on the field may be small due to the findings (of small effects); hence this may be better as a technical note. Additionally, some of the explanation needs improved clarity and some corrections are required, as discussed below.*

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Specific Comments: The importance and impact of this work may be limited for a few reasons, making me question whether this would be better as a technical note than a full paper. The findings indicate that numerical dispersion is less sensitive to the new coagulation formulation/algorithm than to the resolution of the aerosol size discretization. The differences are also much smaller than the simulation errors (determined by comparison to measurements).

20 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

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

We have also added a section featuring a 0D validation of the scheme, in an idealized box setting. It illustrates the benefit of the algorithm in terms of numerical accuracy for different size resolutions.

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 distribution of particles corresponds to the sum of three lognormal distributions of the hazy case of (?). 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 (??) are initialized with concentrations of $2 \cdot 10^{-2} \mu\text{g}\cdot\text{m}^{-3}$. 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 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.

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.

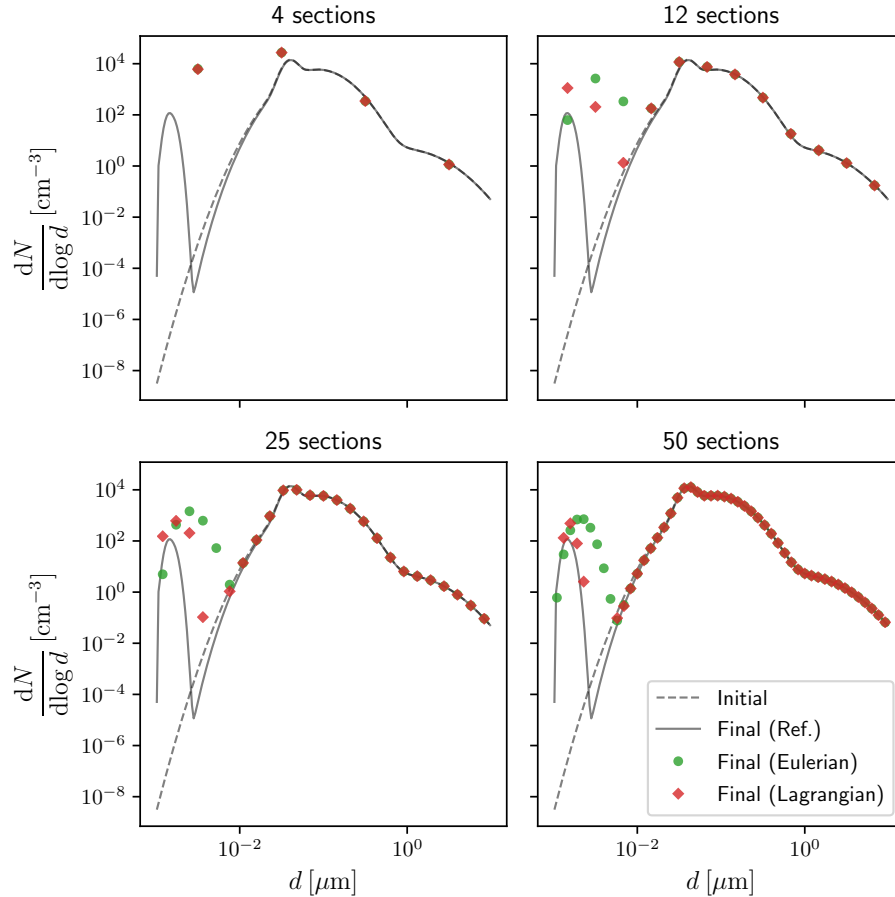


Figure 1. Evolution of the number concentration simulated with the Eulerian and Lagrangian schemes at different size resolutions. The reference is computed with the Eulerian scheme using 200 sections.

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 concen-

tration), 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).

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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 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.

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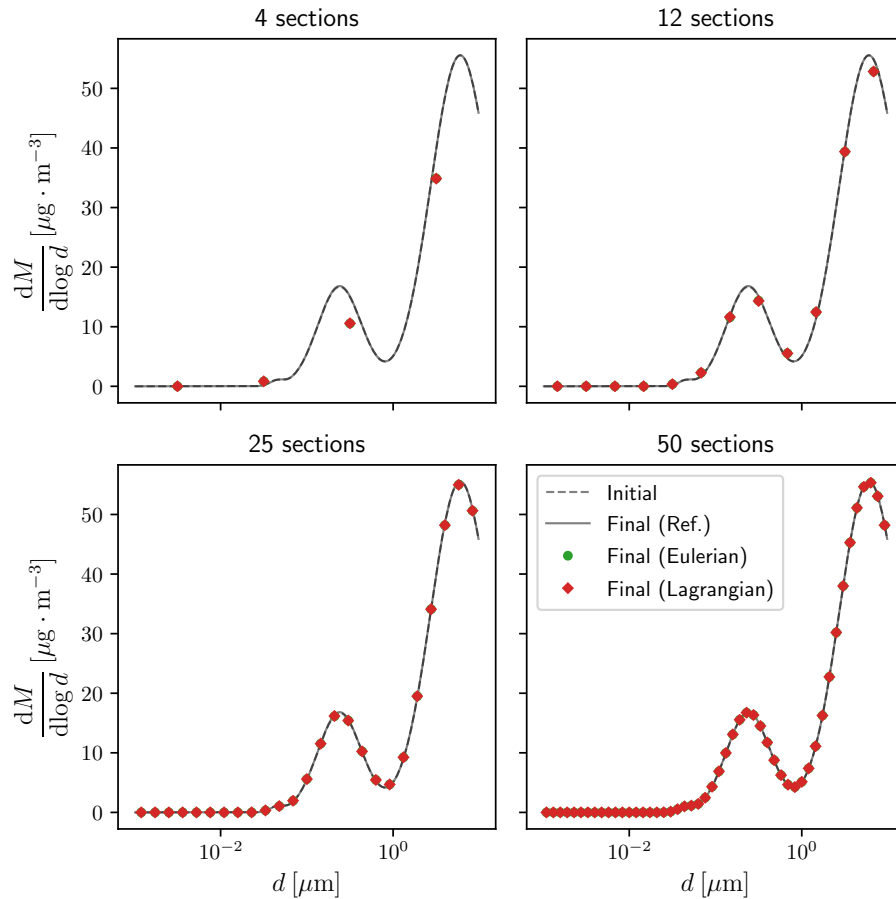


Figure 2. Evolution of the mass concentration simulated with the Eulerian and Lagrangian schemes at different size resolutions. The reference is computed with the Eulerian scheme using 200 sections.

Resolution	Relative error on number concentration (absolute value)					
	$d \leq 10$ nm		$d > 10$ nm		Total number	
	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.

Resolution	Relative error on mass concentration (absolute value)	
	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.

The authors state that the new formulation is basically repeating a derivation of Debry and Sportisee (2007) but correcting a mistake from that paper. This seems like a focus for a technical correction rather than for a novel contribution.

Our reply: Our scheme provides a consistent coupling between aerosol processes in a fully Lagrangian fashion. One step of this scheme consists in an update of coagulation repartition coefficients at every timestep, to account for the evolution of the underlying mesh. This Lagrangian approach was not the focus of Debry and Sportisse (2007), who proposed a closed form for the coagulation partition coefficients regardless of coupling with other processes. During our study, we have noticed that the final closed form reported was inaccurate. Since the evaluation of a closed analytical formulation of these coefficients is key for the overall accuracy and efficiency of our scheme, we have chosen to provide the corrected closed form in Section 2.1, and added a detailed derivation in Appendix C and D.

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The authors further conclude that the new formulation/algorithm may be most useful for low size resolution simulations (9 bins), due to the computational costs of the new Lagrangian formulation/algorithm. Given this, it is not clear why one should invest (computationally) in the new formulation versus just investing in a higher resolution size distribution. A discussion of the relative computational costs should be provided.

Our reply: The objective of the 3D investigation was assessing the relative importance of different error sources. As such, computational time was not the main focus. However, 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.. This remains a valuable trade-off in cases such as the one displayed for the 0D validation, which is not dominated by other error sources which dilute the effectiveness of the Lagrangian scheme. The discussion of the relative computational costs is added in section 2.3.:

95 "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."

100 *Tables 1 and 2 show comparison of model results to measurements for the mixed Eulerian/Lagrangian algorithm (1), not the fully Lagrangian algorithm (2). The text indicates this is because statistics "are very similar". Because the fully Lagrangian algorithm is the focus of this paper, the statistics resulting from using that algorithm should be shown (and perhaps those for the mixed algorithm put in an appendix for comparison)*

Our reply: We have updated Tables 1 and 2 to provide validation statistics about the Lagrangian implementation in the main body of the article. We transferred the statistics about the mixed implementation in an additional Appendix, for comparison.

The Lagrangian statistics are as follows

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

while the Eulerian statistics are given by the following tables, moved to an Appendix

Table 5. 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 Eulerian 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	4806	62	-8	35	8804	7045	99	-20	30
14 sections	5215	5463	92	10	35	8804	8144	99	-7	28
25 sections	5215	5422	92	9	35	8804	8225	99	-7	28

Table 6. 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 Eulerian 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	8.5	94	-12	32
14 sections	10.4	8.7	94	-11	31
25 sections	10.4	8.9	94	-10	30

105 *Additional checking of the derivations is needed (by someone whose work involves similar derivations). I did not have the time to go through the equations in detail to fully understand them, but I did compare them to equations in the existing literature and was left with some questions. First, Appendix A lists the general aerosol dynamics equations for evolution of the number and mass distributions; a citation for this classical formulation is listed as Gelbard et al. (1980) [L91]. Although the number density equations seem generally consistent with other sources (Seinfeld and Pandis, 2006 textbook), I did not find*

110 *these formulations in the Gelbard reference, so the citation seems to need correction. More importantly, it looks like there may be a typo or error in the mass distribution equation (A4) as the units don't seem to work out. Specifically, the last term in the equation ($Is*n$) appears to be missing some kind of multiplicative density term (mass of species s per volume of species s). Without that, there is no mass, so it is unclear how that term contributes to a mass density.*

Our reply: During development, we have checked that our new scheme did provide consistent results with the previously used

115 iterative method. This method was replaced by the closed form, because of its numerical efficiency. The derivation is carried though in less than 10 successive equalities to arrive to a closed form with detailed guidance through the main steps.

We have included Seinfeld and Pandis (2012) as a more general reference which features the general dynamics equation, and included the missing specific density term in equation (A4).

$$\begin{aligned} \frac{\partial n}{\partial t} \Big|_{c/e} (v, t) &= -\frac{\partial}{\partial v} (I_0 n) \\ 120 \quad \frac{\partial q_s}{\partial t} \Big|_{c/e} (v, t) &= -\frac{\partial}{\partial v} (I_0 q_s) + I_s \rho_s n \end{aligned}$$

Additional explanation of some terms is also needed. For example, what is the meaning of the blackboard bold 1 symbol in Equations 3 and 4? Perhaps this is a well-known symbol in this subfield, but I didn't recognize it. If this work is to reach an audience that isn't excessively narrow, non-standard mathematical symbols should be defined.

125 Our reply: We denote by $\mathbb{1}_\Omega$ the indicator function of the domain Ω , meaning it has value one when evaluated within the domain and zero outside. We clarified this notation by adding a definition of this term where it is first used in the manuscript. Equations (3) and (4) are now introduced as

$$\begin{aligned} \frac{dN_i}{dt} &= \frac{1}{2} \sum_j \sum_k N_j N_k \iint dv du K(u, v-u) \mathbb{1}_{[v_{j-1}, v_j]}(u) \mathbb{1}_{[v_{k-1}, v_k]}(v-u) \\ &\quad - \sum_k N_i N_k \iint dv du K(v, u) \mathbb{1}_{[v_{i-1}, v_i]}(v) \mathbb{1}_{[v_{k-1}, v_k]}(u) \\ \frac{dQ_{i,s}}{dt} &= \sum_j \sum_k Q_j N_k \iint dv du K(u, v-u) \mathbb{1}_{[v_{j-1}, v_j]}(u) \mathbb{1}_{[v_{k-1}, v_k]}(v-u) \\ &\quad - \sum_k Q_i N_k \iint dv du K(v, u) \mathbb{1}_{[v_{i-1}, v_i]}(v) \mathbb{1}_{[v_{k-1}, v_k]}(u) \end{aligned}$$

130 with $\mathbb{1}_\Omega$ the indicator function of Ω , such that $\mathbb{1}_\Omega(v) = 1$ if $v \in \Omega$ and $\mathbb{1}_\Omega(v) = 0$ if $v \notin \Omega$.

Table 2 caption. The units of mass concentration appear to be mislabeled here as #.cm-3, which are units for a number concentration, not a mass concentration. The units need to be corrected (or clarified).

Our reply: We have modified the unit of mass concentration to micrograms.m-3. On Table 2, one now reads

135 Mean observed (\bar{o}) and mean simulated (\bar{s}) daily mass concentrations are reported in $\mu\text{g.m}^{-3}$.

Figure 2. The figure labelling should be improved. In addition to labelling the sites based on whether they measure number or mass concentration, sites should also be labelled using the network names provided in the text (LHVP, SIRTA AIRPARIF). Additionally, labeling the location of Paris and the meaning of the polygon's boundaries would be helpful.

140 Our reply: We have labeled individually each measurement station in Figure 2 on a zoomed-in version of the original map, towards the area where measurement station are concentrated. We also specified that background lines represent administrative department boundaries. Figure 2 caption now reads :

Location of observation sites, for reported number and mass measurements. Left panel represents the whole domain considered, right panel represents the area nearest to Paris where most observation sites are concentrated. For geographical context, background lines indicate borders of administrative departments around Paris area, the most central one indicating the city of Paris.

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Technical corrections: Figure 3. The exponent (4) is missing from the color scales and the units of number concentration are also missing (except for the exponent).

Our reply: Thank you for pointing this out. We have also noticed additional missing characters in other figures, in labels as well as legends and graduations. After careful verification on our part, it seems that problem only came up after files were transferred to the journal. Those issues are not present in the original figures, which are also available on the Zenodo archive provided and look as intended. We will try to resolve this issue with the editor, as it seems to be purely rendering related and this aspect is not reproducible on our part.

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155 *L20 typo: should be “focused” L42 “formerly equivalent to ...’ I don’t understand this. Should it say “formally”?* L86. *Missing word “be” before “a partitioning” L107 Typo: “substraction” L186. Should refer to Figure 6, not Figure 3.*

Our reply: We have updated the manuscript with the corrections proposed by the referee, thank you for pointing them out.