AC Response to RC1

We thank Alexei for taking the time to review our manuscript and appreciate the constructive feedback provided.

The secondary ice production (SIP) mechanisms receive exceedingly more attention in cloud research, as their importance in atmospheric ice formation becomes evident. There is, however, a strong disbalance between modelling efforts, field research, and laboratory experiments dedicated to the elucidation of SIP mechanisms, with the modelling studies clearly dominating the research landscape. Whereas in-situ aircraft-based research was recently gaining momentum thanks to new cloud particle imaging probes becoming available, the experimental efforts are scarce and are not yet capable of delivering physically meaningful and quantitative description of the SIP mechanisms so badly needed by cloud modelling community. Quite understandably, the SIP modelling scientists cannot wait for very slow advance of the state-of-the-art experimental activity and are therefore dependent on sieving the experimental data from past literature sources or venturing an extrapolation from recent but very preliminary experimental data.

The manuscript by R. James et al. reports on the modelling of various SIP mechanisms contributing to the enhancement of ice crystal number concentration (ICNC) in shallow convective clouds featuring cloud top temperature above -10°C. The main focus of the study lies in evaluation of the secondary ice formation upon collision between a rain droplet (diameter more than 150 µm) and an ice particle resulting in "splashing" of the droplet. In this mechanism, called "freezing fragmentation of droplets – mode 2" and subsequently referred to as "M2", the secondary water droplets produced in such a splashing event may contain fragments of ice crystals forming at the moment of droplet freezing initiation upon contact with ice. This mechanism was recently suggested on a basis of theoretical considerations in Phillips et al. (2018) and was not seriously considered being relevant for SIP, until the recent study of James et al. (2021) which produced some evidence of its potential importance. My comments are therefore mostly focused on the parametrization and implementation of this mechanism, because this is where my doubts are the strongest.

General comments.

RC: A. According to (Phillips et al., 2018), the main criterium for M2 being active is the value of dimensionless energy DE that has to be higher than the critical value DE_{crit} (eqs. 7 to 9 of the manuscript). If this is the case, secondary droplets are produced upon collision of water droplet with solid ice surface (splashing condition). This criterium is accepted in the study as a basis for parametrization of secondary ice production rate according to M2 mechanism. (Phillips et al., 2018) derived this criterium analysing the results of droplet-droplet collision reported in (Low and List, 1982) and using the dimensionless collision energy formalism of (Testik et al., 2011), hinting at the absence of other laboratory data.

I seriously doubt that the critical value of DE obtained by applying the formalism of (Testik et al., 2011) to the droplet-droplet collision data can be immediately used for parameterizing splashing due to droplet-solid surface collision.

AC: As stated in Phillips et al. (2018), observations of drops on rough copper hemispheres from Levin and Hobbs (1971) were also used to constrain the Mode 2 splashing parameterisation and the available data from Levin and Hobbs (1971) also fit the parameterisation. Admittedly there are not many data points, but the data we have fit the parameterisation.

RC: Actually, this is also not the most straightforward approach as there is quite a number of studies describing splashing of droplets upon collision with curved solid surfaces. In many studies, a combination of dimensionless numbers (Weber, *We*, and Ohnesorge, *Oh*) is used to describe the onset of splashing when the critical collision velocity is reached for the droplet of given diameter. For example, (Charalampous and Hardalupas, 2017) have found that splashing of water droplets colliding with a larger spherical target is initiated for *We* larger than 400, whereas this value is around 120 for droplets colliding with the flat surface. Sykes et al. (2022) has used a more complex splashing parameter based on a combination of kinetic energy of collision, surface energy, and viscosity, but even there splashing does not occur for We less than 300.

AC: Both Charalampous and Hardalupas (2017) and Sykes et al. (2022) show that the splashing onset is related to the Weber number. The dimensionless energy (DE) is related to the Weber number as shown below except by a factor of 12. The results from Charalampous and Hardalupas (2017) and Sykes et al. (2022) therefore validate the general theoretical approach used in Phillips et al. (2018) for parameterising Mode 2. Charalampous and Hardalupas (2017) have not formulated an SIP parameterisation, so we have resorted to using the Phillips et al. (2018) formulation.

RC: Sykes et al. (2022) also contains a nice overview of previous experimental results on droplet-surface collisions and is definitely worth discussing. Apparently, many factors influence the splashing behaviour upon collision, including roughness of the solid surface, amount of water adsorbed on the surface, and the curvature of the surface. And yet, none of these factors are included into the M2 parameterization of (Phillips et al., 2018).

AC: We agree that roughness of the solid surface ought to affect the splashing behaviour. Roughness is not included in the M2 parameterisation, but then again there are no cloud microphysical models, that we are aware of, that calculate the roughness of the ice particle surfaces. Therefore, it does not help to include roughness in the parameterisation.

RC: It is straightforward to compare the critical Weber number with the DE introduced by Phillips et al. (2018), because *DE* is just the so called modified Weber number (the ratio of kinetic energy of collision to the surface energy):

$$We = \frac{\rho_w \cdot D_{drop} \cdot v_{col}^2}{\gamma_l}, DE = \frac{\rho_w \cdot D_{drop} \cdot v_{col}^2}{12 \cdot \gamma_l} = \frac{We}{12}$$

Here I take for simplicity that the droplet is much smaller than the ice particle and therefore the mass term in equation (7) is just the mass of the droplet m_d. If the droplet and the ice particle are approximately of the same mass, then $DE = \frac{We}{24}$. From here one can easily define the relationship between the droplet size D_{drop} and the relative collision velocity v_{col} required to initiate splashing (see Figure 1 below). One immediately notices that according to splashing criteria based on $We > 120$ (collision with flat surface, the red dash line) the relative collision velocity for a given droplet size must be almost 10x higher than the velocity estimated from the condition of $DE_{crit} = 0.2$ (the blue dash-dot line). It is even worse if the condition of $We > 400$ must be satisfied to initiate splashing. The argument that the splashing was observed in the experiments reported by James et al. (2021) remains valid, because the collision conditions were indeed well above the splashing limit (the solid diamond data point at $D_{drop} = 5\ mm$ and $v_{\mathrm{col}} = 5.3\ m/s$).

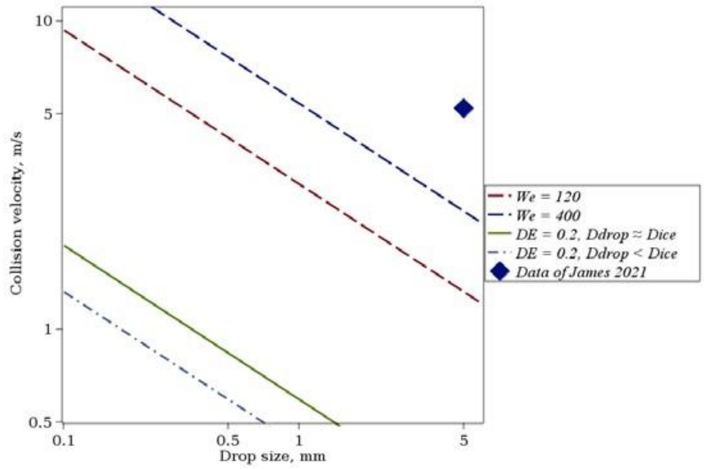


Figure 1. Splashing condition for droplet-solid surface collision.

So, the central question becomes, would the M2 mechanism contribute to any secondary ice production if the condition of $We > 120$ or even $We > 400$ has to be fulfilled to ensure splashing upon collision? Could you extend the sensitivity study into this range or is it rather pointless because neither droplets nor ice crystals grow to the sizes required to fulfil this condition?

AC: Data from Levin and Hobbs (1971) was used to constrain the M2 parameterisation of Phillips et al. (2018). Levin and Hobbs (1971) specifically designed their experiments to represent cloud conditions. The amount of water adsorbed on the surface and roughness were accounted for by using a dry, roughened copper surface and the curvature between the drop and target had a ratio of 10 %, not untypical for clouds. These results are implicitly factored into the Mode 2 parameterisation of Phillips et al. (2018).

The issue with saying that We > 300 or 400 must be fulfilled to ensure splashing are the experimental conditions in which this value was determined. Charalampous and Hardalupas (2017) used smooth particles composed of glass to reduce the *'influence of the surface roughness… thus reducing the complexity of the interpretation of the measurements'*. Sykes et al. (2022) used *'smooth untreated borosilicate-glass substrates'*. Ice in clouds is unlikely to be as smooth as the substrates in Charalampous and Hardalupas (2017) and Sykes et al. (2022) and there is no reason to take their critical Weber numbers over the roughened substrates of Levin and Hobbs (1971).

However, we do accept that there is large uncertainty in the DE_{crit} value, which is influenced by all of the factors mentioned above and have performed sensitivity studies on the DEcrit value by increasing it by a factor of 10 and 30. The results are given in Sections 4.1.4 and 4.2.4 for a natural and near-city aerosol size distribution respectively.

4.1.4 Sensitivity test: M2 DE_{Crit}

The M2 parameterisation given in Eq. 9 requires the onset of splashing, denoted as DE_{crit}. We used the value of 0.2 given in Phillips et al. (2018) based on laboratory data of drops colliding on roughened copper hemispheres from Levin et al. (1971). To test the sensitivity of ice enhancement to DE_{crit} during M2 simulations, we ran simulations for warmer and colder cloud bases with natural aerosol size distribution and updraft speeds of 2 m s^{-1} using the following DE_{crit} values: 0.2, 3 & 6. The results are plotted in Figure 5.

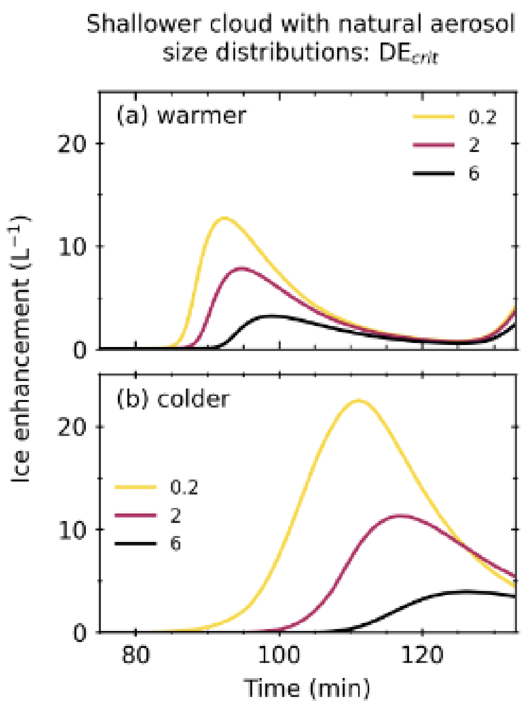


Figure 5. M2 ice enhancement against simulation time for three DE_{crit} values (0.2, 3 and 6) for a shallower (1.3 km) cloud with natural aerosol size distributions and updraft speed of 2 m s^{-1}. Warmer refers to cloud base temperatures of 7 °C, and colder refers to cloud base temperatures of 0 °C.

4.2.4 Sensitivity test: M2 DE_{Crit}

Figure 9 shows the M2 ice enhancement against simulation time for three DE_{crit} values, 0.2, 3 and 6, for shallower clouds with a near-city aerosol size distribution. In general, lower values of DE_{crit} had earlier maximum ice enhancement peaks compared to higher values of DE_{crit}. The maximum ice enhancement peaks were greater for lower DE_{crit} values.

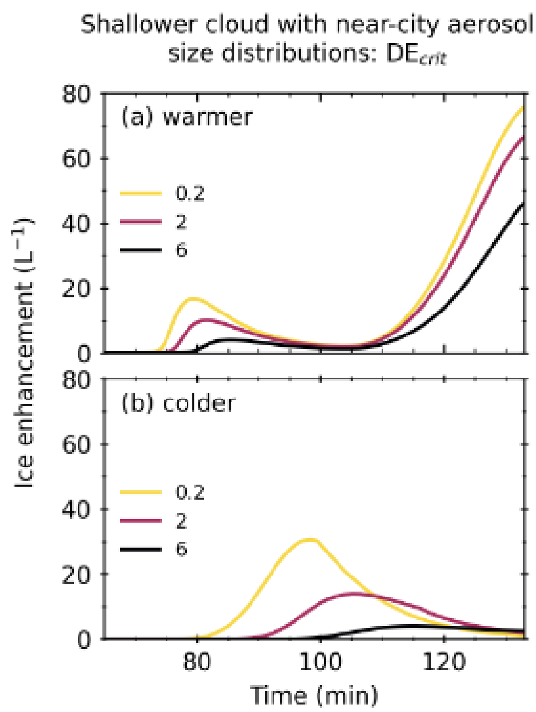


Figure 9. M2 ice enhancement against simulation time for three DE_{crit} values (0.2, 3 and 6) for a shallower (1.3 km) cloud with near-city aerosol size distributions and updraft speed of 2 m s^{-1}. Warmer refers to cloud base temperatures of 7 °C, and colder refers to cloud base temperatures of 0 °C.

We have also expanded the discussion of DE_{crit} to the following:

Other uncertainties in the M2 parameterisation include the critical value for onset of splashing (DE_{cit}). For splashing to occur, the dimensionless energy must be greater than DE_{crit}. Phillips et al. (2018) estimated a value for DE_{crit} of ~0.2 based on room temperature experiments of colliding drops by Low and List (1982) and acknowledged that this was a source of uncertainty. The equation used to describe number of secondary liquid drops was further constrained using laboratory experiments by Levin et al. (1971). These experiments consisted of 2.5 mm drops impacting on a rough copper sphere at room temperature. The number of secondary drops formed in collision experiments is sensitive both to the geometry and impact material; hence the expression used to describe the number of liquid fragments presents another source of uncertainty. We increased the DE_{crit} by a factor of 10 and 30 to demonstrate the sensitivity of M2 ice enhancement on the DE_{crit}, as

shown in Figs. 5 and 9. When the DE_{crit} was increased by a factor of 10 or 30, the ice enhancement decreased compared to the stated DE_{crit} in Phillips et al. (2018), but ice enhancement still occurred. Despite the uncertainties within the M2 parameterisation, and in agreement with Phillips et al. (2018), it is more erroneous not to include M2 within simulations. Our laboratory experiments strongly suggest that the M2 SIP mechanism could be active (James et al., 2021), and further laboratory studies into this mechanism will help reduce the uncertainties listed.

B. Examination of the Figure 1 brings some follow-up questions. If the less restrictive condition of $DE > 0.2$ can be used, what are the collision velocities that can be achieved assuming the free fall of droplets and ice crystals? Since the bin model handles the collisions explicitly and the significant enhancement of ICNC due to M2 is achieved, the collisions are obviously efficient in the model; however, it would be very useful to show the distribution of sedimentation velocities for droplets and ice particles as a function of cloud evolution. I would be especially interested to see the sedimentation velocities of ice particles with the aspect ratios and perhaps even effective densities predicted by the ice growth model. A comparison of the modelled sedimentation velocities for both ice particles and rain droplets with the values observed in the real shallow convective clouds would also be very instructive.

AC: Although the bin model treats the collisional speeds, it has not been output in the model simulations and it is not straightforward to us how to display this. We also did not observe sedimentation velocities in the real clouds so do not feel this would be particularly instructive. We used Pruppacher and Klett (2010) to calculate sedimentation speeds of droplets over the whole range. The main aim of the paper was to try to show, given what we currently know about SIP, how does the 'mode 2' parameterisation behave in a cloud model simulation compared to other SIP mechanisms. We feel that the findings in the paper show that it is potentially very important, and that it warrants further quantification of the mechanism. There are outstanding questions, which are yet to be addressed, but can be the subject of further study.

C. I was surprised to learn that apparently, this study is not the first attempt to include M2 into the cloud modelling. As mentioned in the Discussion section (lines 556 to 569) Zhao et al. (2021) and others have modelled the contribution of M2 mechanism using essentially the same parameterization provided by (Phillips et al., 2018). However, no significant contribution to SIP has been demonstrated. So much is clear from the text of this manuscript but the detailed discussion of the possible reasons is missing.

Given that the M2 is in the focus of this study, I feel that the manuscript would strongly benefit from such discussion.

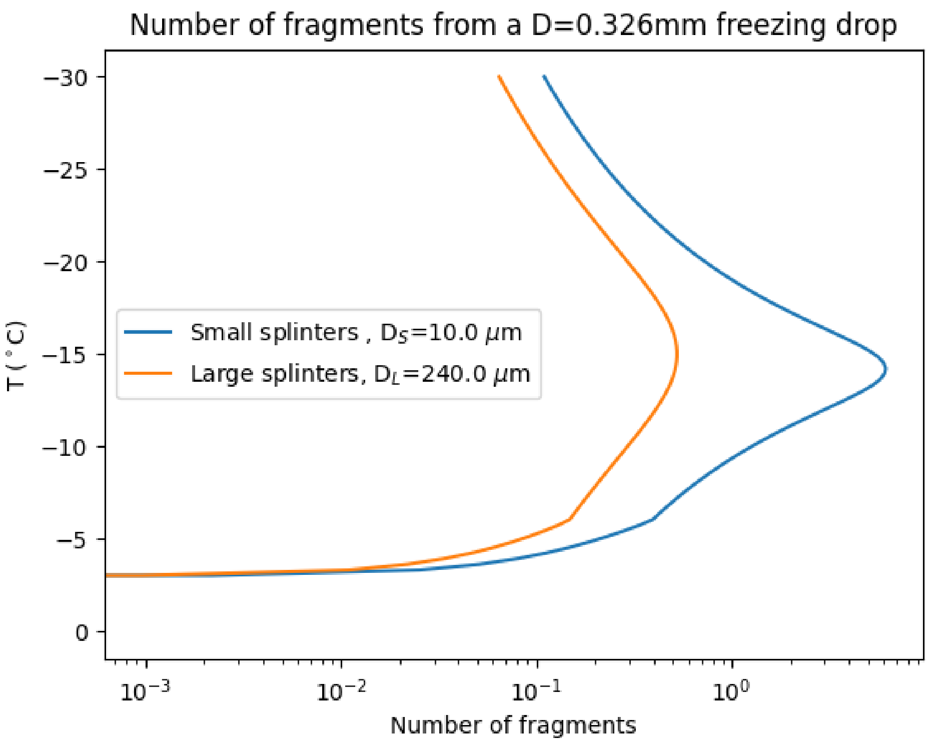
AC: The original phrasing of the paragraph gave the misrepresentation that M2 did not significantly contribute to SIP. We have rephrased so that it is clear that for some studies M2 did contribute significantly, but M1 may have had a larger contribution. We have also expanded the discussion to include reasons given from the studies at to why M1+M2 were not significant.

Several studies have included M2 in their simulations, although usually combined with M1 (Phillips et al., 2018; Sotiropoulou et al., 2020; Qu et al., 2020; Zhao et al., 2021; Zhao and Liu, 2021; Georgakaki et al., 2022; Zhao and Liu, 2022; Huang et al., 2022; Karalis et al., 2022) which is equivalent to our M1+M2 simulations. For our idealised cloud conditions, the contribution from our M1+M2 simulations appears to be derived from the M2 aspect. In fact it is only when we combine M1 with M2 that we see any significant ice enhancement from M1. On its own, M1 was not a strong ice enhancement mechanism in our idealised shallow convective clouds. In contrast, Phillips et al. (2018) (see their Fig. 6) showed that in their simulation of a deep tropical maritime convective cloud with temperatures between 0 to -20 °C, over 90 % of the ice came from M1 and M2, of which M1 contributed to approximately double that of M2. M1 was more significant in Phillips et al. (2018) simulations compared to M2, probably due to simulation temperatures which covered the thermal peak of M1 at -15 °C. As shown in Fig. 10, very few fragments were formed away from the thermal peak. Most of our simulations were warmer than the thermal peak and the deeper clouds with colder cloud base temperatures of 0 °C only briefly had temperatures near the thermal peak early on in the simulation (see Fig. S2(b) of the Supplementary). Similar results were also observed by Qu et al. (2020) who modelled similar deep tropical maritime convective cloud conditions that Phillips et al. (2018) simulated. Zhao et al. (2021) simulated four types of Arctic mixed-phase clouds based on Atmospheric Radiation Measurement Mixed-Phase Arctic Cloud Experiment observations. They showed that approximately 80 % of all ice particles came from M1 in their single-layer boundary layer stratus simulations. The largest contribution of M2 came from the transition simulations but this only contributed a small fraction compared to M1. Other studies which modelled the M2 SIP mechanism with the M1 SIP mechanism did not provide a breakdown of the contribution from M1 and M2 (Zhao and Liu, 2021, 2022; Huang et al., 2022; Karalis et al., 2022). Two studies found that M2 combined with M1 was not an effective SIP mechanism in the simulated conditions (Sotiropoulou et al., 2020; Georgakaki et al., 2022). Sotiropoulou et al. (2020) stated that this was due to their thermodynamic conditions with relatively cold cloud base temperatures and Georgakaki et al. (2022) stated that this was due to the drops being too small to initiate M1 + M2 in their simulated conditions.

D. On a side note, a few experimental studies of the freezing fragmentation of droplets (M1 mode) under more realistic conditions of free fall has been published after 2018 (Kleinheins et al., 2021; Keinert et al., 2020; Lauber et al., 2018). It might be worth checking the parameterization of (Phillips et al., 2018) against the data of these recent studies.

AC: We acknowledge that there have been several interesting studies on the freezing fragmentation of droplet (M1). However, the selection criteria of the laboratory data used in the Phillips et al. (2018) mode 1 parameterisation specifically states that it only included studies where the drops were in free fall. Thus excluding Keinert et al. (2020) and Lauber et al. (2018), both of whom used an electrodynamic balance to levitate drops. We note that Keinert et al. (2020) simulate free fall by passing an airflow around the drops. We also note that Keinert et al. (2021) shows that compared to Lauber et al. (2018), having an air flow around the levitated drop clearly enhances breakup. Therefore, checking the parameterisation of these new studies against Phillips et al. (2018) mode 1 parameterisation needs to be done carefully, and we feel this is outside the scope of our paper.

Kleinheins et al. (2021) present pressure release events rather than number of SIP particles because it is difficult to see small splinters visually. However, they find 3x more pressure release events than ejected particles (counted visually in tKeinert et al. [2020]). Keinert et al. (2020) found that typically a thermal peak at -15 °C occurred where there were around 1-2 events per droplet. The number of small splinters from the Phillips et al. (2018) parameterisation for the same-sized drops is shown below. This has a peak at -15 °C of around 6-7, which is the same order of magnitude as Kleinheins et al. (2021); however, Kleinheins et al. (2021) showed that the peak was not centred on -15 °C. Although there are differences, it is not clear which is correct – as acknowledged by Kleinheins et al. (2021), the number of pressure release events do not necessarily equate to the number of SIP particles for instance. Given the uncertainty we feel it is justified to use the parameterisation and comment on the state of the science in the paper.



Summarizing, I find the presented study a useful exercise in modelling the SIP contributions to ice enhancement in a shallow convective cloud and as such well worth publishing. Not being a part of modelling world myself, I can only presume that the model framework itself is flawless and can handle the standard microphysics properly, given the validity of underlying parameterizations. In case of M2, I have strong doubts that the splashing condition is fulfilled in a shallow convective cloud and that the M2 SIP mechanism will be important under the circumstances. I do not doubt, however, that if the splashing condition were fulfilled, the M2 would become a valid SIP mechanism, but the validity of parameterization should be convincingly shown, through experiments or via small-scale physical modelling of a collision-freezing event. One possible suggestion would be to choose the modelling case differently (deep convective cloud?) to reproduce these conditions, but this would be a different study. I feel this is a major critical point since the study is dedicated to the evaluation of the M2 mechanism and therefore request “major revisions”.

AC: See above responses.

Minor comments:

RC: Line 73: Reference to (Hobbs and Rangno, 1990) is given twice.
AC: Removed.

RC: Table 1: why is the effective density of *larger* colliding particle and the aspect ratio of the *smaller* colliding ice particle is given? Aren't both parameters for both colliding particles are important to calculate the relative collision velocity?

AC: It is not the relative collision velocity that is being calculated here. Relative collision velocity is calculated from the difference in sedimentation speeds in the model, which depends on the density and aspect ratio as mentioned. Table 1 is used to identify the parameters of the surface of the ice category, which go into equation 13 of Phillips et al (2017). It is to be used in conjunction with Table 1 of Phillips et al (2017)

RC: Equation (9) should include $\max\,(DE - DE\ \ , 0)$ as in (Phillips et al., 2018), to avoid getting negative values for subcritical dimensionless energy (if $DE < DE\ \ $).
AC: Changed.

RC: Line 156: $f(T)$ is the mass FRACTION of water converted to ice after the end of the first freezing stage, not the "mass of frozen drop…".
AC: Changed.

RC: Figure 4: the line colours for Φ values of 0.3 and 0.1 should be swapped. The way it is shown, the lower value of Φ results in a stronger ice enhancement.
AC: Changed.

RC: Supplement, figures S17 to S20: The colour bar on the right supposedly gives the number of particles per unit volume; the units of the number concentration are not given.
AC: Added units.

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