**Response to Reviewer 2**

We thank the reviewer for providing insightful comments and suggestions on our paper. We believe that responding to the comments has further improved the quality of the manuscript. We have addressed the comments below. The reviewer comments are in **black**, our responses in **red** and any text changes in **blue**.

**Review 2**

This work quantitatively investigated the evolution of an evaporating droplet and its surrounding environment utilizing an idealized numerical model. With different settings of ambient temperature, relative humidity, pressure, and initial droplet radius, this work examines the evolution of droplet temperature and lifetime and their dependences with these environment factors. The findings confirmed the previous literature that assumes steady-state droplet temperature, and the main novelty is finding that droplets can be much colder and last longer, due to the cooling of the adjacent air and the temperature gradient in the immediate environment surrounding the droplets. The results of this study is particularly of interest to the modeling community, which has been struggling with the underestimation of INPs for a long time. I think this manuscript is generally well written and recommend its publication in ACP, with some comments listed below.

Detailed comments:

1. Line 31: what do you mean by "cells"? do you mean a grid in numerical models?

   By "ice-generating cells", we refer to small convective cells at tops of otherwise stratiform clouds. The American Meteorological Society's Glossary of Meteorology defines "generating cells" as "In radar, a small region of locally high reflectivity from which a trail of hydrometeors originates". The term was first used in 1953 by J.S. Marshall (Marshall, J. S., 1953: Precipitation trajectories and patterns. J. Atmos. Sci., 10, 25–29, doi:10.1175/1520-0469(1953)010<0025:PTAP>2.0.CO;2.). A good review of generating cells can also be found in Plummer et al., 2014, included in our reference list.

   To avoid any further ambiguity, we have now added a reference to the AMS Glossary definition in the revised text.

2. Line 70-72: this sentence has too many sub-sentences, suggesting rewrite it.

   We have simplified the text to read as follows:

   Lü et al., (2005) conducted ice nucleation experiments with acoustically levitated supercooled water droplets. Using statistical analyses of nucleation rates, they found that ice nucleation predominantly initiates in the vicinity of the droplet surface.

3. Line 151: I am curious if there are any differences between using cylindrical coordinates and spherical coordinates in the model, as the droplet volume, surface curvature and water tension may be calculated differently in the two coordinates.

   We repeat our response to Reviewer 1 who also had similar concerns.

   We have expanded the description of the coordinate systems that are used in COMSOL. For 3D problems, COMSOL has an option to use cartesian, spherical or cylindrical coordinates. For a 2D

axisymmetric domain used in this study, the default spatial coordinate system used by COMSOL is cylindrical coordinates, which in 2D is the same as a cartesian coordinate system in r and z. To avoid any further confusion, we have removed the word cylindrical and used cartesian coordinates instead.

The physics and the governing equations are the same irrespective of coordinate systems, with the only difference is that all dependent variables (temperature, vapor concentration, etc) will be expressed as functions of r and z. The form of differential operators such as gradient and divergence also changes accordingly. The built-in interface takes care of these changes, along with applying the boundary conditions at appropriate boundaries as indicated. More details regarding the coordinate transformations can be found here. In the end, the results will be independent of coordinate system chosen.

It is actually more complicated since COMSOL uses three coordinate systems simultaneously (referred to as spatial, material and mesh frames). We have added additional information in the text (Section 2.1) to help the reader understand the Arbitrary Lagrangian-Eulerian (ALE) framework utilized in this study to track the moving boundary of the shrinking droplet. The revised text is also given below:

The simulation of the spatiotemporally varying droplet temperature and radius of an evaporating cloud droplet embedded in a gaseous domain is difficult to solve analytically because of the moving and shrinking boundary at the surface of the evaporating droplet. These kinds of moving boundary problems are also known as Stefan problems. To model this process, we have used an advanced numerical solver, COMSOL (Version 6.0), which employs a finite element method to solve partial differential equations (PDEs). The COMSOL Multiphysics software simultaneously uses spatial, material, and mesh coordinate systems described as the spatial frame, material frame, and mesh frame, respectively. The spatial frame is a fixed, global, Euclidean coordinate system, which in 2D has spatial cartesian coordinates (r, z) with the center of the droplet at (r, z) = (0,0) (Fig. 1). The material frame specifies the material substance, in this case, water or air. The mesh frame is a coordinate system used internally by the finite element method.

The Navier-Stokes and Fick's second law of diffusion equation, which follows from the continuity equation, along with appropriate boundary conditions (see Sec. 3) are solved to conserve mass and momentum in the whole system. The following physics interfaces in COMSOL were used to simulate droplet evaporation: (1) *Two-Phase Laminar Fluid Flow*, which includes a moving mesh to track the shrinking water–air interface of the evaporating water droplet and fluid–fluid interface that incorporates evaporative mass flux; (2) *Transport of Diluted Species* to track water vapor diffusion through the air domain and predict the evaporation rate at the droplet surface; and (3) *Heat Transfer in Fluids* which accounts for the non-isothermal flow within the computational domain, temperature-dependent saturation vapor density at the droplet interface, and a boundary heat source to account for the latent heat of evaporation. The computational domain also includes an infinite element air domain (COMSOL 2023b) to specify and maintain boundary conditions far away from the droplet. The physics modules are coupled through non-isothermal flow between heat transfer and fluid flow, and mass transport at the fluid–fluid interface between fluid flow and species transport.

A non-uniform moving mesh was created by breaking down the computational domain into numerous fine elements of variable sizes, using the Arbitrary Lagrangian-Eulerian technique (Yang et al., 2014) to accurately track the moving air-water interface at the droplet surface. In the ALE technique, the spatial cartesian coordinate system (r, z) is fixed, while the coordinates of the material (R, Z) and the mesh ($R_m$, $Z_m$) nodes are functions of time as the droplet evaporates. However, the material and mesh node coordinates are always fixed in their respective frames. Initially, the spatial, material and mesh frames are all identical. As the simulation starts, the material and mesh frames

deform as the moving boundary of the droplet shrinks during evaporation. After each time step, the deformed nodes are mapped to the spatial frame, where calculations are performed. In this study, we have used triangular mesh elements (COMSOL 2023c) within the droplet and quadrilateral mesh elements (COMSOL 2023d) for the rest of the domain as shown in Fig. 1. The triangular mesh allows a higher resolution at the droplet surface, and both meshes adjust continually as the droplet surface shrinks during evaporation. Finally, to simulate the water droplet evaporating in ambient air system, with appropriate initial and boundary conditions, the discretized PDEs are numerically solved with adaptive time steps (≤ 0.01 s) to maintain numerical stability and obtain the solution (the temporal evolution of droplet temperature and radius) for a range of conditions.

4. Line 170-171: Can you add a few sentences describing why it uses different meshes in and out of the droplet? any pros and cons for this setting?

   The triangular mesh allows a higher resolution at the droplet surface and the mesh adjusts continually as the droplet surface shrinks during evaporation. We have now added this information to the revised paper.

5. Line 190-191: maybe change the temperature unit from K to C for easier read. Same as the figures and tables.

   We have considered this option but decided to keep all the temperatures in Kelvin for ease of comparison with our previous paper (Roy et al., 2023). In addition, regeneration of all the figures and tables and text would be difficult in the given timeframe to submit the revised paper.

6. Line 211-213: related to my comment #3, maybe this is something can be used to explain that using cylindrical coordinates is appropriate.

   Please see our response to Comment #3.

7. Line 252: give a number to the equations.

   All equations are now numbered.

8. Line 257: μ should have a value, what is the number?

   COMSOL uses temperature dependent empirical formulae for the dynamic viscosity of water and air. However, for water below 273.15 K, the dynamic viscosity is approximated as 1.79 mPa s. For air, the equation is an empirical equation that produces values equivalent to Sutherland's law.

   We have added this information to the revised paper.

   "….and $\mu$ is the fluid dynamic viscosity. For water below 273.15 K, the dynamic viscosity can be approximated as 1.79 mPa s. For air, COMSOL uses an empirical equation that produces values equivalent to Sutherland's law (White, 2006), $\mu = \mu_0 \left(\frac{T}{T_0}\right)^{\frac{3}{2}} \left(\frac{T_0 + S_\mu}{T + S_\mu}\right)$

   where $\mu_0 = 1.716 \times 10^{-5}$ N s m$^{-2}$, $T_0 = 273$ K, and $S_\mu = 111$ K for air. The empirical equation is given as:

   $$\mu = -8.38278 \times 10^{-7} + 8.35717342 \times 10^{-8} T - 7.69429583 \times 10^{-11} T^2 + 4.6437266 \times 10^{-14} T^3 - 1.06585607 \times 10^{-17} T^4 \qquad (4)$$"

9. Line 268: k should be a constant or function depending on T and p, what is the number? And is it different in the droplet and in the environment air?

The value of thermal conductivity of water used in the simulations is constant at 0.56 W/(m K). We chose this value based on Fig. 3 of Biddle et al, (2013) where the thermal conductivity of supercooled water is very close to 0.56 W/(m K) for the range of temperatures in the simulations. We have added information and a reference to Biddle et al., (2013) in the revised paper.

Based on Beard and Pruppacher (1971), the thermal conductivity of air has very weak dependence on temperature over the temperature range used in the simulations. Their thermal conductivity ($k_a$) equation is given by

$$k_a = 0.004184[5.69 + 0.017(T - 273.15)] \quad (\text{W m}^{-1}\text{ K}^{-1})$$

For both T = 273.15 K and 253.15 K, the value of $k_a$ is 0.02 W m$^{-1}$ K$^{-1}$. Hence, we have used a constant value of 0.02 W m$^{-1}$ K$^{-1}$. We have added this information to the revised paper:

We chose the value of $k$ for supercooled water at 0.56 W m$^{-1}$ K$^{-1}$ based on Fig. 3 of Biddle et al., (2013) where the thermal conductivity of supercooled water is very close to 0.56 W/(m K) for the range of temperatures used in this study. Based on Beard and Pruppacher (1971), the thermal conductivity of air, given by $k_a = 0.004184[5.69 + 0.017(T - 273.15)]$ (W m$^{-1}$ K$^{-1}$), has very weak dependence on temperature over the temperature range used in this study. For both T = 273.15 K and 253.15 K, the value of $k_a$ is 0.02 W m$^{-1}$ K$^{-1}$. Hence, we have used a constant value of 0.02 W m$^{-1}$ K$^{-1}$.

10. Line 276: one factor that impacts the final temperature drop at the droplet surface is the difference of water diffusivity and heat diffusivity of the environment air. I am wondering how large the diffusivity uncertainties of water and heat are, and how this will impact the temperature drop.

We tried to find information on uncertainties in these values. Biddle et al, (2013) doesn't discuss uncertainties. Unfortunately, we were unable to find information the reviewer requests. We can assume that that the uncertainty is no larger than the last significant figure reported for these values, which suggests very high accuracy given the number of significant figures reported.

11. Line 292: $T_\infty$ should be in the unit of K when multiplied with R.

The reviewer is correct and that was what was done. However, the way the sentence was phrased was misleading. The sentence originally read:

For the vapor transfer interface, except within the droplet, all domains are at an initial vapor concentration of $c_{0,air}$ which is again assumed to be the same as that of the constant ambient concentration value far from the droplet, $c_\infty$, calculated as follows:

$c_\infty = \frac{RH_\infty \times e_{sT_\infty}}{R_{univ} \times T_\infty}$ where, $RH_\infty$ is set at a constant ambient relative humidity far from the droplet, $R_{univ}$ = 8.3145 (J/mol/K) and saturation vapor pressure, $e_{sT_\infty} = 610.94 * \exp\left(\frac{17.625 * T_\infty}{T_\infty + 243.04}\right)$ (in Pa, with $T_\infty$ in °C) following Alduchov and Eskridge (1996).

We have now changed the sentence in the revised paper to read:

For the vapor transfer interface, except within the droplet, all domains are at an initial vapor concentration of $c_{0,air}$ which is again assumed to be the same as that of the constant ambient concentration value far from the droplet, $c_\infty$, calculated as follows:

$c_\infty = \frac{RH_\infty \times e_{s_{T_\infty}}}{R_{univ} \times T_\infty}$ where, $RH_\infty$ is set at a constant ambient relative humidity far from the droplet, $R_{univ}$ = 8.3145 (J/mol/K), $T_\infty$ is in K. The saturation vapor pressure is calculated as, $e_{s_{T_\infty}} = 610.94 * \exp\left(\frac{17.625*T_\infty}{T_\infty+243.04}\right)$ (in Pa, with $T_\infty$ in °C) following Alduchov and Eskridge (1996).

12. Line 309: again, T should be in the unit of K here.

The reviewer is correct and that was what was done. The way the sentence was phrased was misleading. The sentence was changed in the revised paper to read as follows:

Hence, saturated vapor concentration at the shrinking droplet boundary, using the ideal gas law, is given by, $c_{sat}(T_{sf}) = \frac{e_s(T_{sf})}{R_{univ} \times T_{sf}}$ where $T_{sf}$ is the surface temperature, in K. The saturation vapor pressure $e_s(T_{sf})$ is estimated as $e_s(T_{sf}) = 610.94 * \exp\left(\frac{17.625*T_{sf}}{T_{sf}+243.04}\right)$ (in Pa, with $T_{sf}$ in °C) again following Alduchov and Eskridge (1996).

13. Line 396: the number of mean cooling rate (K/s) is huge but does not mean anything, it is just an initial model spinup. Maybe just remove it.

Thank you for the suggestion. We have removed all instances of mean cooling rate.

14. Line 399-401: These numbers are different from the numbers in Figure 4.

The numbers in the original Figure 4 (now Fig. 3) for final temperature of the droplets were averages of the three values for $r_0$ = 10, 30 and 50 µm because it was hard to fit all three numbers on the figure. We have changed the caption to make that clear. The caption now reads,

Droplet temperature evolution (left column) and radius evolution (right column) for three different $RH_\infty$ ($RH_\infty$ = 10% (brown curves), 40% (orange curves) and 70% (green curves)), three different $r_0$ ($r_0$ = 10 µm (dot-dashed lines), 30 µm (solid lines) and 50 µm (dashed lines)), with three different $T_\infty$ = 273.15 K (0°C) (a, b), 268.15 K (-5°C) (c, d) and 263.15 K (-10°C) (e, f), for $P$ = 500 hPa. For each $RH_\infty$, the average droplet temperature at the end of the lifetimes of the three droplets with different $r_0$ ($T_L$, in K) is given in (a,c,e) and the time taken to reach the end of its lifetime ($t_L$, in s) is given in (b, d, f). Exact values of final temperature for each $r_0$ are given in Table 1.

15. Figure 4: again, I suggest using C instead of K for the unit of temperature. This makes y axis cleaner.

As discussed earlier, we had considered this option but decided to keep all the temperatures in Kelvin for ease of comparison with our previous paper (Roy et al., 2023). In addition, regeneration of all the figures and tables and text would be difficult in the given timeframe to submit the revised paper.

16. Section 4.4: The presentation in this section needs to be improved. The authors list many numbers for different conditions, easily making readers get lost which quantity is in comparison (e.g., Section 4.4.2). I strongly suggest the authors simplify the text. For example, saying that "For environment with RH=10%, T=273K, P=500hPa, the lifetimes of 10, 30, 50 um diameter droplet are 1.1s, 1.4s,

32.8s, respectively." (well, the effect of droplet size to lifetime is obvious, maybe section 4.4.2 can be removed or modified).

In response to this comment and reviewer 1's concern, we have reduced the number of figures by eliminating the three figures showing the results at $P$ = 850 hPa since the results are summarized in Tables 1 and 2. We have expanded our discussion of the remaining figures and the tables in Section 4 to focus on the physics behind the results.

17. Table 1: again, using C instead of K makes it easier to read.

Please see our previous response (Comment 5).

18. Table 2: I would not put lifetime difference ($t_L$ - $t_{LC}$) in the table, or just use a relative difference (percentage change), which is more relevant to the modeling application.

Thank you for the suggestion. We have added two extra columns to Table 2 that show the percentage changes.

19. Line 592-605: This paragraph may need to be re-organized or re-stated. It currently reads like saying the previous assumption of steady-state droplet temperature is imperfect and this study improves it. However, this study verified that the steady-state droplet temperature assumption is valid, with the main novelty to be considering the gradient of adjacent environment, which was not considered in previous studies.

We tried to soften the paragraph, so it didn't give the impression that we are criticizing past work. We have removed the phrase "In order to model a more realistic scenario of an isolated droplet evaporating in a subsaturated environment". We note that our study showed that the steady-state droplet temperature assumption is not valid for conditions where droplets are evaporating in environments with moderate to low relative humidity. In very high relative humidity environments, approximately > 90%, the steady-state assumption can provide a reasonable estimation of the droplet temperature.

To be clear, we showed that the steady-state droplet temperature assumption is not always valid. This can be directly seen by comparing the $T_{RRD}$ column (the steady-state temperature) with $T_L$ column (temperature from the current simulations) in Table 1. For the range of ambient relative humidities we simulated (10-70%), the droplet temperature deviates from the steady-state simulations. For example, for 50 µm droplet, for an ambient temperature of 263.15 K, the temperature deviation from the steady state-solution can vary from 2.6 K for $RH_\infty$= 70% to 16.2 K for $RH_\infty$ = 10%. Even for the smaller deviation, the impact on potential ice nucleation events can be significant, because of the strong dependence of ice-nucleation rates on temperature. As noted in response to the first reviewer, at very high relative humidities (e.g. $RH_\infty$ = 99%) the steady-state assumption is valid.

20. Line 623: I am curious whether the RH = 10% is realistic in real world. In another words, do we really have a droplet ~ 25 K colder than we thought?

In situations where air has a history of descent above a stratiform cloud deck, the vertical humidity gradient can be quite large. Observations of relative humidity vertical profiles above Arctic stratocumulus clouds reveal strong vertical gradients in RH at cloud top, with RH values as dry as ~ 50% (Egerer et al., 2021). Also, during the SNOWIE field campaign, dry layer incursion (with RH ~10-20%) was observed above orographic clouds, which led to very sharp gradients in RH at cloud

tops (Xue et al. 2022). Here, in this study, we have chosen 10% and 70% to bound the simulations on both ends.

Egerer, U., Ehrlich, A., Gottschalk, M., Griesche, H., Neggers, R. A. J., Siebert, H., and Wendisch, M.: Case study of a humidity layer above Arctic stratocumulus and potential turbulent coupling with the cloud top, Atmos. Chem. Phys., 21, 6347–6364, https://doi.org/10.5194/acp-21-6347-2021, 2021.

Xue, L., and Coauthors, 2022: Comparison between Observed and Simulated AgI Seeding Impacts in a Well-Observed Case from the SNOWIE Field Program. J. Appl. Meteor. Climatol., 61, 345–367, https://doi.org/10.1175/JAMC-D-21-0103.1.

21. Line 633: it also includes 10 μm droplet.

Thank you for pointing this out. We have now added the 10 μm droplet to the sentence in the revised paper.