Referee comment by Gabor Vali on "Retention During Freezing of Raindrops, Part I: Investigation of Single and Binary Mixtures" by Gautam et al.

This manuscript extends to raindrops the work previously done by the authors and their colleagues on retention coefficients for cloud droplets. Acoustic levitation in a cold room and chemical analyses before and after freezing constitute the essence of the experiments. The levitation system and the use of infrared thermometry avoid the need for contact with any support. This is a near ideal arrangement. The paper present a good description of the experiments and sound analyses of the retention coefficients. The paper is well constructed and well written (with a few odd phrasing). The topic is quite appropriate for ACP.

This reviewer has not been involved for decades with the topic of retention of foreign material from ice growth and will only address in these comments the physical aspects of the experiments, how to interpret the results, and to what extent the results apply to processes in atmospheric clouds.

Two features of the experimental approach are the focus of these comments: the large difference in temperature between the drop and the surrounding air, and the near-absence of ventilation.

The 2-mm diameter raindrops used in the experiments are up to 100 times larger than the cloud droplets used in the previous experiments and thus have about 100 times larger volume to surface ratios. That would lead one to anticipate significantly slower freezing and, consequently, larger rejection of foreign substances as ice forms. The results here presented show the opposite. For two of the substance involved in both experiments (formic and acetic acids), values near 0.7 were obtained in the riming experiments and near 1.0 for the raindrops. The authors' chief argument for this is that the larger drops in free air had an ice shell form on their outside trapping most of foreign substances.

The formation of the ice shell after nucleation is well documented in the paper. It is also what one would expect for an isolated drop with the air temperature considerably lower than the drop temperature even before nucleation and pronouncedly so during the freezing of the drop when the surface temperature rises to near 0°C (Fig. A1). In contrast, in the atmosphere, the temperature of the drop would be close to the air temperature before freezing. It would also have asymmetric heat transfer when nucleation and initial ice formation leads to latent heat release within the drop. The resulting surface temperature and the formation of ice within the drop will be influenced by the asymmetry and by the rate of heat transfer to the environment. Theoretical analyses of the problem have been made with respect to hailstone formation and growth (e.g. List, 2014). These analyses also consider evaporation from the drop surface and collection of cloud droplets, but do not treat explicitly how ice forms inside the drop. For the current discussion, more relevant are the many experiments, and drops caught in clouds, that demonstrate that frozen drops often have protuberances and other deformation on their surfaces. Cracks in the ice shell may lead to the expulsion of liquid to the surface and perhaps to the air. The theory of ice multiplication in clouds by splintering is based on those observations (Field et al, 2017; Lauber et al., 2018).

The potential for cracks in the ice shell may also have to considered for the experiments described in the paper. Internal pressure rises as the drop freezes and is likely to produce cracks in the ice shell (e.g. Korelev and Leisner, 2020; Kleinheins et al., 2021; references herein). Because of the low air temperature in the experiments, any excluded water is likely to freeze onto the surface quite rapidly. This would slow internal freezing. The cited papers describe work with water without added substances. Dissolved gases or ions may modify the freezing behavior.

Most of the foregoing work was done with droplets of hundreds of micrometer in diameter, not far but still below the size of 2 mm involved in the current experiments. That discrepancy and the complex nature of the phenomenon make any extrapolation difficult and it is even more speculative how all of the above influence retention of foreign substances. In that light, it is a welcome development to have the results presented in this paper. However, it is clear that more work is needed and that the authors of this paper should express their views on the matter in the manuscript.

Another dimension of the problem is how the high retention found in this work might be envisaged on the molecular scale. Some discussion of the results of molecular simulations of crystal growth may help readers' understanding of the results.

Section 3.5 deals with the retention indicator defined by the relative timescales of mass expulsion and that of freezing. It is unclear if this measure is intended to describe and idealized freezing front or is applied to specific geometries, spheres in this case. Perhaps the authors can illuminate this by justifying their choose of the parameters used to calculate the retention indicator. Specifically, the choice of the time of ice shell formation as the freezing time needs justification.

Unless the points raised in the foregoing can be shown to be unimportant, the Conclusion section should include less definite statements about complete retention in clouds.

Minor points:

Unless already well embedded in the literature, the terms "riming-retention" and "freezing-retention" should be reconsidered. The latter could apply to both riming (small droplet) and raindrops. The 'droplet' vs. 'drop' distinction is generally accepted in the literature and although imperfect as a definition it may be better to use the terms 'retention in freezing droplets' and 'retention in freezing drops'. Unfortunately, while it would be useful, it is impractical to also include in the terms some indication of what is being retained. Maybe acronyms have to be relied on.

line 18-20: suggest using " ...aerosols from the boundary layer .." and " ... troposphere, and that can alter ...."

line 58: suggest 'visualize' instead of 'conceptualize' line 62-62: suggest to replace 'infer a more systematic understanding' with a simpler 'improve understanding'

line 67: omit 'which was'

Eqn (3) might add the explicit result combining (1) and (2). Also would be informative to get some idea of the magnitude of D for the experiments for different temperatures.

line 143 and others: it would better to avoid the phrase 'freezing profiles' as there are too many different contexts for freezing already. Perhaps 'temperature graph' or just 'temperature] could be used. Even less useful is 'INP freezing profile'.

References:

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Lauber, A., A. Kiselev, T. Pander, P. Handmann, and T. Leisner, 2018: Secondary ice formation during freezing of levitated droplets. Journal of the Atmospheric Sciences https://doi.org/10.1175/JAS-D-18-0052.1.

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