

We would like to thank the Editorial Support team for giving us the opportunity to respond to the reviewers' comments in open discussion. We greatly appreciate the reviewer's careful reading and review of this manuscript. We have addressed each reviewer's comments and suggestions, as shown in our responses below. Specific references to the manuscript have been highlighted in yellow and rewritten sections are provided directly below in italics for convenience. The lines, figures, and sections referenced match the numbering in the version of the manuscript presently uploaded for discussion. A revised version of the manuscript will be uploaded for the handling editor's consideration.

Reviewer(s)' Comments to Author:

RC1:

The manuscript is based on extensive laboratory studies on the mechanism of retention/release of semi-volatile organic compounds upon freezing of raindrop-sized droplets generated from the aqueous extracts of Beijing wintertime urban aerosol. The whole set of experiment is carefully designed and executed, using state-of-the-art equipment including analytical techniques, and the statistical processing of analytical data is adequate meeting all standards of science. The objectives of the manuscript are clear, the hypotheses are valid and important, to explain a potential transport mechanism of semi-volatile organic compounds to the free troposphere. This is relevant in understanding new particle formation in the upper troposphere, which has serious implication on cloud formation and water-particle interactions in a changing climate hosting more water vapour in the atmosphere. The manuscript is comprehensive and well-written, so there is little to criticize expect typography (e.g. using 'en dash' characters instead of the 'minus sign' in all formulae.) However, the reviewer has a series of serious concerns about the relevance of the laboratory results for real-life atmosphere.

We would like to thank the reviewer for the constructive comments. The typography issues concerning the use of 'en dash' characters and 'minus sign' characters have been addressed where found. We request the editorial team review our compliance with the journal's formatting guidelines on this issue as appropriate.

- **In the study water-soluble organic compounds (WSOC) in the aqueous extract of wintertime urban aerosol serve as a proxy for real-life composition of droplets of convective clouds. However, large-scale convective cloud formation is not typical during the winter, furthermore, in winter frequent inversion and low mixing layer height prevent surface emissions to be transported to higher**

altitudes and participate in ice cloud formation. Whereas I agree that studying such a complex mixture may be more informative than that of a few cherry-picked model species, to draw meaningful conclusions from the experiments it is important to elaborate on this issue in the manuscript.

We have incorporated discussion of this relevant concern in **section 4, line 511**. We would like to address that our filter samples were chosen to maximize mass loading, which was mainly achieved in the winter months.

It is also important to note that large-scale convective cloud formation where freezing retention is expected is not typical during the winter such as when the aerosols in the present study were sampled. Furthermore, in winter, frequent inversion and low mixing layer height tend to prevent surface emissions from being transported to higher altitudes and participate in ice cloud formation, so the compounds presented here may not be wholly representative of the species that participate in the retention process.

- **The laboratory experiment is designed to study the freezing of large (2 mm in diameter) raindrops. In real-life mixed phase clouds freezing of large supercooled droplets may occur via multiple mechanisms, such as riming, immersion or contact freezing. These processes are also active for much smaller cloud droplets, i.e. for those below or around the precipitation threshold. I wonder what the probability is for such a large (supercooled) raindrop to survive such effective freezing processes inside a vigorous mixed phase cloud high in the troposphere? I would guess it is very low, but it would be worth discussing anyway.**

For a large, supercooled raindrop to survive such conditions is low. This might be possible in a thunderstorm cloud, but more relevantly, the chosen drop size is rather an experimental constraint. First, a relatively large liquid mass is required for chemical analysis. Second, in order to properly determine the freezing temperature, a certain drop diameter is needed. So in order to ensure viable measurements, we were required to scale the process to the larger drop sizes one might find in the atmosphere. For small drop sizes, freezing is less relevant than riming.

Riming is established as the dominant pathway for ice production in convective systems, but hetero/homogenous freezing is still an understudied contributing pathway. Previous studies have already begun to examine retention during riming. These experiments use large wind tunnel facilities to simulate rime growth. One of the objectives of this study is to establish untargeted complex mixture analysis as a method to measure retention before

we take it to the wind tunnel. We have future experiments planned as a separate publication involving the Mainz wind tunnel facility to more closely examine rime growth.

- **In addition, the real-life freezing processes described above are superfast relative to the cooling process applied by the authors in their experiments, which lasts on average for 90 seconds. So again, the question arises how relevant the experimental parameters are for real-life cloud conditions in determining the gas-to-droplet partitioning of organic compounds?**

While the total freezing was approximately 90 seconds, the most relevant freezing is likely the ice shell formation, which is very quick, on the order of milliseconds. Freezing time as a relevant physical parameter for the freezing process is addressed more in-depth in the Part I publication. <https://doi.org/10.5194/egusphere-2024-3917>

- **Another issue to be clarified is the application of the standing ultrasonic wave for levitating the droplets in the experimental setup. It is well-established that ultrasonic energy is an extremely effective way of mixing (see e.g. ultrasonic bath for extraction). So in terms of fluid dynamics, can we assume that in the laboratory experiments the droplets had remained thoroughly mixed until they froze up? If so, how it relates to fluid dynamics prevalent for droplets in convective clouds, in which mixing inside such large droplets may be way much less effective?**

We do assume that the ultrasonic field produces droplets that are well-mixed. Before they stabilize, the drops perform several visible oscillations that produce mixing. There is a more thorough investigation of drop mixing in the acoustic trap in Szakáll et al., JAS, 2009 (<https://doi.org/10.1175/2008JAS2777.1>). However, we would also say that for a 2 mm raindrop in the atmosphere that falls with 6-7 m/s, turbulent mixing will occur inside the drop. While mixing should be higher in an ultrasonic field, mixing inside a freely falling drop is also high and therefore it is justifiable that our drops are also well mixed before freezing.

Ensuring homogeneity inside the droplet is also important for maintaining reproducibility for this experiment. The calculations we use for determining freezing retention assume the droplet is at chemical equilibrium before freezing. Nonhomogenous conditions inside the droplet would invalidate these assumptions. However, we estimate that it would not produce significantly different results specifically for freezing retention so long as diffusive influence could be exactly controlled and accounted. While nonhomogenous mixing of large droplets would relevantly influence diffusive transport as it creates a concentration gradient, its influence on the freezing retention should be negligible. As a phase separation process, unless a species is near to saturation concentrations (unlikely in naturally

occurring scenarios and would likely be insoluble and unretained to begin with), the freezing retention has not been demonstrated to be concentration dependent.

- **The last issue is that in large droplets mass transfer to the atmosphere is strongly limited by the low surface area to mass ratio. Furthermore, freezing of the droplets starts from the outside because the enthalpy of fusion needs to be dissipated, then freezing forms a solid outer layer that hinders heat and prevents material transport to and from the interior of the droplet. I wonder if this mechanism is largely responsible for the nearly complete retainment of soluble species irrespective of the vast range of their physical and chemical properties?**

We appreciate the reviewer's good observation. This 'ice shell' formation is discussed is addressed more in-depth in the Part I publication. <https://doi.org/10.5194/egusphere-2024-3917>

We would like to add that the retention can also be understood as the ratio of the chemical expulsion timescale and the freezing time (or ice shell formation time).

To summarise my general comments it would be more than welcome if the authors addressed these points in their revised manuscript.

We would like to thank the reviewer for their time and their contribution of constructive comments.

RC2:

This study presents the retention of organics during the freezing of rain size droplets. The experiments were conducted on aqueous extracts of Beijing urban ambient aerosols using an acoustic levitator. The retention coefficients were determined by freezing the rain size droplets and then measured the remained substances in the frozen droplets. They showed near 1 of retention coefficients for the compounds found in the HRMS in (-)HESI mode, but more variation for the compounds found in (+)HESI mode. Correlations between the estimated chemical properties, such as henry's low constant, and retention coefficients were discussed. The subject of this manuscript fits the scope of ACP. There are several issues need to be addressed before it can be considered for publication.

We would like to thank the reviewer for the constructive comments.

Major comments:

- 1. The manuscript presented in the current form is more like a Measurement Report, as its atmospheric implication is not fully discussed or addressed. The only three nighttime samples also limits a broader atmospheric implication. If I understood correctly, the samples were combined before the freezing and determination of retention coefficient. Why not perform the experiments for individual samples, which one can exam the variation of retention for the same compounds?**

There are a variety of reasons why we chose to combine filter samples for our experiment. Using combined samples instead of individual samples eliminates variability to allow for experimental reproducibility and valid statistical evaluation of the retention coefficients. Specifically, some of these confounding variables that are controlled by this method could be large concentration differences between samples, background matrix influences, pH, biasing of trace species, etc. The individual samples on their own also do not produce adequate mass loadings for experimental viability. Combining sample extracts allows for enough liquid volume to be produced for the experiment without unduly diluting the sample. Further, comparative analysis of the individual filter samples is outside objectives of this publication and would also require examining the synoptic conditions during filter sample collection.

- 2. In the Abstract, the statement in Line 20-22 is overstated as the S- and N-containing compounds are not solely from the NO_x and Sox chemistry. The statement in Line 24-26 is not supported by the results of non-sigmoidal relationship or the related discussion. The lower surface-to volume ratio of the large drop size investigated may be one of the reasons. The differences in the experimental techniques, as I understand, may be the main reason, the wind tunnel and levitation, which have totally different air dynamic environments and surrounding corresponding gas-phase organic concentrations that will affect the gas-liquid diffusion and partitioning, and thus the retention.**

We have amended **lines 20-22** to more accurately state what compounds are being referenced. We have amended **lines 24-26** to more precisely state the confidence of correlation. We agree that the differing physical parameters of this experiment is a significant barrier to its comparison with wind tunnel studies; we present discussion on this at the **end of section 3.3** and in the conclusions in **lines 469-474**. One of the objectives of this study is to establish untargeted complex mixture analysis as a method to measure retention before we make more direct comparisons to the wind tunnel. We have future experiments planned as a separate publication involving wind tunnel experiments. With

interest for this discussion, our preliminary data from the tunnel shows similar results to this experiment in the levitator. This gives us reason to believe that the physical differences of these experiments is not the cause for the absence of sigmoidal behavior here.

The findings here also indicate that N- and S- containing compounds, likely secondary organic aerosols (SOA) anthropogenically related to NO_x and SO_x chemistry, have enhanced retention. An insignificant positive correlation between polarity and freezing retention along with a significant negative correlation with vapor pressure and freezing retention was observed.

- 3. The limitation of this technique or the limited number of sample size should be discussed. For example, as the authors indicate the implication for the convective clouds, will the values measured by this levitation techniques be likely underestimated for the convection system?**

We have updated **line 120** to highlight the actual number of drop samples that were collected and analyzed from this experiment (30 drops). We have expanded our discussion in **section 4, particularly lines 512-517** to better address the limitations and implications of this study to convective clouds.

Enough drops to reach the minimum viable sample volume for analysis, 50 µl, were collected to produce a single sample (approximately 12 drops). 30 drops were produced in total, which allowed for two full samples to be used for UHPLC-HRMS analysis.

It is also important to note that large-scale convective cloud formation where freezing retention is expected is not typical during the winter such as when the aerosols in the present study were sampled. Furthermore, in winter, frequent inversion and low mixing layer height tend to prevent surface emissions from being transported to higher altitudes and participate in ice cloud formation, so the compounds presented here may not be wholly representative of the species that participate in the retention process.

- 4. The correlation of retention coefficients with chemical properties. The discussion on the results of these correlations should be carefully examined. For example, Line385-392, the R square values are less than 0.1 and the F-test shows no significance, why do the statements still say that they have correlations? Then, the related conclusions are not valid anymore.**

For the precision of statistical discussion, an insignificant correlation is still considered a correlation. To state that there is no correlation is to demonstrate that the population correlation is zero. Our failure to reject the null hypothesis however does not confirm the null hypothesis. The r-squared and F-test indicate that there is no practically important linear relationship, not that there is no linear relationship, i.e. that population correlation is zero. The F-test indicates that we can't prove r is significantly different from zero. It doesn't prove that r is 0 with significance. However, we agree that this phrasing can be misleading, especially outside of statistical context. We have adjusted the phrasing used in throughout **section 3.3**, specifically **lines 385-395, 411, 430, and 433** to swap some instances of 'correlation' with 'regression' as to not imply undue significance. The conclusions around the insignificant correlations found are still valid.

The determined molecular weight (MW) shows little correlation linearly with freezing retention (as seen in Figure S2). In (-)HESI, the slope of the regression is weakly positive, suggesting larger compounds are more likely to be retained. This is likely related to lower vapor pressures associated with larger MW species in the negative mode. An F-test against the constant value model indicates that this correlation is not significant (p-value: 0.0857). However in (+)HESI, the slope of the regression is weakly negative for MW which suggests the opposite. An F-test against the constant value model indicates that this correlation with MW is also not significant (p-value: 0.1440). This trend in the positive mode is likely driven more by polarity, as Figure 1.A also demonstrates that larger species in (+)HESI tend to have higher HPLC retention times and are therefore more nonpolar. The plot in Figure 7 further demonstrates this with a stronger negative correlation between the HPLC retention time and the freezing retention

Other comments:

- 1. Line 227-234, it is not clear what are those compound number means. How many compounds were used for and analysis? Does that include the 77 and 84 compounds which were selected for additional property calculation?**

The MS features described in **lines 227-234** are relevant to understanding how the untargeted mass spectrometry dataset was handled. **Lines 228 and 232** state directly how many compounds were used for analysis from the positive and negative modes. From those 208 and 250 compounds, the 77 and 84 were then used for the additional property calculations.

- 2. Line 235-237, the sentence is confusing.**

We would like to thank the reviewer for the constructive comments. We have rewritten **lines 235-237** for clarity.

Phosphorous containing species can represent almost a third of positively ionizable species in rainwater WSOC (Seymore et al., 2023) so these species likely make up a significant portion of species variety that is not considered.

3. Figure 2B, why there are such a large portion of the compounds have retention coefficients higher than 1.0?

Retention coefficients greater than 1.0 are an indication of a certain level of experimental error. Since the retention coefficient is measured relative to a standard, any change in the signal of the standard will falsely inflate or decrease the measured retention coefficient.

4. Line316, why a nonnormal distribution is expected or is a true distribution?

A normal distribution may indicate that all retention coefficients are a constant value with a normal distribution of error. We expect a nonnormal distribution as this would indicate that we are observing a range of retention coefficients, as we expect from a variety of species with different H^* .

5. Line393, in the sentence, the Figure 8 should be Figure 7?

We have corrected this typo in **line 393**.

6. Line398 and Figure 8, can you comment on why the R square is so low?

An F-test against the constant value model is a comparative test that indicates whether the proposed regression is more likely to be true than the null hypothesis, i.e. no correlation. If the F-test passes, we can state that the correlation (Pearson R) is not zero even if the R^2 is low. It indicates that these chemical properties are likely factors in determining retention (Pearson R is not zero), but they alone cannot fully explain retention (R^2 is low).

7. Line492 and 505, the authors mean “indicates” not “insulates”?

We have fixed these typos in **lines 492 and 505**.

8. Line505, lower potential to retain, why is likely to reach the upper atmosphere?

Species that are less retained are believed to be in the gaseous state after freezing and therefore able to make it to the convective outflow. This is in contrast to a species that is retained in the hydrometeor and then deposited in wet deposition. These freshly oxidized SOA precursors described have higher VP and therefore are less likely to be retained than aged or primary organics.

We would like to thank the reviewer for their time and their contribution of constructive comments.

RC3:

This paper addresses an important phenomena potentially impacting new particle formation in the upper troposphere, retention during raindrop freezing. The work substantially expands the breadth of compounds considered previously through the use of well-described state-of-the-art methods and sophisticated analyses. Results are interesting and provide insights into differences in retention based on compound classification and functionality. They further suggest impacts of atmospheric chemical processing on retention. However, the discussion of theory-based hypotheses and rational is somewhat lacking and leaves out relevant previous work, particularly regarding the expected impacts of freezing conditions on retention of low Henry's law constant compounds. Further, the conclusions on retention of general chemical classes and the impact of NO_x chemistry are overstated based on the evidence presented and on the necessary uncertainty associated with large classes of partially identified compounds. They should be more nuanced. The manuscript is generally easy to read and understandable but is sometimes repetitive and vague. It also includes too many graphics that are somewhat repetitive or not informative enough for the main text. Detailed data on the compound data informing the analysis should also be provided in a supplemental table.

We would like to thank the reviewer for their constructive comments. The MS data the reviewer requests is too large to be meaningfully incorporated into a supplemental table and therefore we will provide it through zenodo. We have published the requested data as two .csv files on that platform. <<https://doi.org/10.5281/zenodo.15166745>>

Specific comments, listed by line number (more or less in sequential order):

Abstract (and conclusions): The conclusions on retention are too broadly applied to entire classes of chemicals when there is not enough data specifically for that class. The least supported is for 'sulfides' when only one organosulfide was identified in the dataset. The authors acknowledge this in line 368, but still state the unsupported general conclusion in the abstract and conclusions.

Line 20 and line 475 directly state that these species are the least observed in the dataset. To avoid unintended overstatement, these lines have been reworded to refer more specifically to the observed compounds of that class. Even then, our reasoning in **line 368** still supports that the speculation of low retentions for these classes could be a reason for their underrepresentation in the dataset.

Most nitroaromatics and organosulfates were fully retained along with the aliphatic amines (AA) and higher-order amines and amides while the observed sulfides, lipids, aromatic hydrocarbons, and long chain compounds are among the most unretained and incidentally the fewest species present.

20 and 476. The fact that sulfides, lipids, aromatic hydrocarbons, and long-chain compounds were not observed in high quantities is not incidental to the conclusions of this work, as small sample size was acknowledged as likely impacting the results.

Please see our reply to the previous comment.

21-22. The statement that anthropogenically related NO_x and SO_x chemistry enhances retention of the resulting secondary organic aerosol' is confusingly written. Are the authors saying that the compounds created are more polar and less volatile, increasing the formation of/partitioning to the condensed phase of the aerosol, and also retention of component species during freezing? Clarify this.

We have amended **lines 20-22** to more accurately state what compounds are being referenced.

The findings here also indicate that N- and S- containing compounds, likely secondary organic aerosols (SOA) anthropogenically related to NO_x and SO_x chemistry, have enhanced retention.

23-25. The sentence is a run on sentence and uses a double negative, which is confusing.

We have split this sentence in **lines 23-25** into two separate sentences to increase clarity.

No sigmoidal relationship with effective Henry's law constant was observed. This differs with the parameterizations of riming retention presented in current literature, which is justified by the lower surface-to-volume ratio of the large drop size investigated.

105-113. A diagram of the freezing experimental setup is needed.

The diagram and associated images of the experimental setup are more presented and more thoroughly explained in the Part I publication. <https://doi.org/10.5194/egusphere-2024-3917>

156. Provide a brief reason why phosphorus containing species were not considered.

Naturally occurring phosphorus containing compounds are present almost exclusively as organophosphates. This means that there is always at least 4 O atoms per P atom present. The heuristic filtering necessary to ensure these correct ratios while maintaining compound assignment validity was outside of the scope of the analysis. More specifically, performing accurate compound assignment for P containing compounds is difficult using the native version of CompoundDiscover. It often requires externally processing the data using MIDas or an in-house software. Newer versions of CD are making this process better and more accurate so future studies using untargeted analysis will include P containing compounds.

156 (and elsewhere). “level 5 (L5) and higher compounds”. Based on the context, I think ‘higher’ is referring to L1-4. This is confusing because 1-4 are lower numbers than 5. This is confusing; it would be better to just provide the L range being referenced.

We have incorporated this into the manuscript in **lines 156, 157, 165.**

All level 5 (L5) through level 1 (L1) compounds...

169. “These properties” suggests more than one type of property was predicted here. However, only the effective Henry’s law constant is mentioned. Where other properties predicted here or is this a typo?

There is more than one property that has to be incidentally calculated to estimate an effective Henry’s law constant; specifically vapor pressure and aqueous solubility. It is also calculated through two different methods; bond contribution or group method. For simplicity we omitted these details and were left with the grammatical error. We have amended **line 169** accordingly.

This was predicted using the HENRYWIN™ model as part...

219-220. Provide some rationale for nonuniform desorption effects being negligible.

Desorption is largely dependent on vapor pressure and equilibration time. Our samples are from HiVol filters, which bias for a relatively narrow range of vapor pressures.

330. “The means of the composition classes in (-) HESI vary little, generally less than 5% from each other”. This statement is vague and misleading. The range of means is

0.90 to 0.99 in Table 1. Just state the range rather than making a statement that could misrepresent the data.

We have incorporated this into the manuscript in **line 330**.

The means of the composition classes in (-)HESI range between 0.90 and 0.99 with an unweighted average of 0.95 and a standard deviation between them of 3%. In contrast, the means of the composition classes in (+)HESI vary more, up to 40%.

338. “CHO has lower retention ...than CHNO in both ionizations”. This is not entirely supported by Table 1 for the positive ionization for which CHO had mean retention of 1.01 and CHNO had a lower mean of 0.94. The median is lower for CHO, but not the mean. This needs a more precise and nuanced description.

We have incorporated a more precise description into the manuscript in **line 338**.

Notably, with lower median retentions in both ionizations, most CHO compounds have lower retentions than CHNO compounds.

345-346. The conclusion that nitrogen chemistry of CHO enhances retention is not strongly supported by the data described here, given the above inconsistency (see comment on line 338), and the small differences between the means compared to the error (with error indicated by measured retention coefficients of a lot more than 1 for many compounds). This really should be stated more as a hypothesis of expected differences between composition class and what might explain them, and the degree to which the evidence supports (or doesn't support) the hypothesis.

The previous comment to increase clarity on line 338 does not invalidate the conclusions made in lines 345-346. Both the mean and median for CHO are less than those for CHNO in the (-)HESI. While the mean is higher for CHO than CHNO in the (+)HESI, the variance is significantly larger (0.82 vs 0.21) and the median is lower (0.90 vs 0.93). While that alone gives enough information to state there are more species with lower retention in CHO than in CHNO, the visual distributions in Figures 5B and 6B also demonstrate that the lower two quartiles in CHO are significantly lower than those for CHNO.

Figures 2, 3 and 4 and Tables 2 and 3 could be provided in supplemental materials, as they are not instrumental to the main results.

In our opinion, Figures 2, 3, and 4 as well as Tables 2 and 3 are instrumental to demonstrating that the retention coefficients we are measuring are not simply a normal distribution of error around a constant value. Further, they are necessary for any reader who intends to incorporate these results into a model.

Figures 5 and 6 are not both needed and could be combined into one figure (such as with an overlay, or just eliminate Figure 5)

We believe that Figures 5 and 6 are both necessary and demonstrate independent points. Figure 5 demonstrates the differences between retentions of compound classes in a statistically familiar way. Figure 6 is to demonstrate which compound classes follow nonnormal distributions and which we believe are normal distributions around constant values.

Supplemental material should include a detailed table of compounds used in the analyses (those supporting the tables and figures shown in the main text), including their level, masses, structures, assigned composition, properties predicted, and CAS number match (when applicable and used for properties rather than a model prediction).

Please see our previous reply.

375. The sulphur-containing class had the highest mean/median retentions. This should be mentioned and discussed. Line 492 later concludes that SO_x reactions enhance retention, but this is not discussed with the results.

Discussing heteroatom classes like such is not always practical, as they are composed of compound functional groups with differing properties. Especially with the S- containing species being some of the fewest species observed in the dataset, it was not useful to discuss heteroatom groups as such. The conclusion in line 492 is based on the discussion of organosulphates found in the CHOS (-)HESI from [lines 252-257](#).

385-391. The trends with molecular weight do not seem to warrant this much discussion given they are inconsistent. Further the reasons involve the influence of vapor pressure and polarity, and require the evidence for each of those relationships. I would make more sense to put this paragraph after those discussions (and shortening it).

As requested, we have moved the discussion of MW paragraph from [lines 385-394](#) to after the discussion of polarity ([Lines 395-406](#)). We maintain that this discussion of MW is relevant as it is a fundamental chemical property with direct implication for the polarity and VP of a compound. It is also a directly measured chemical property rather than an estimated property like VP or H*. It would be remiss if it was not properly discussed despite its inconsistency as a predictor of retention.

392-3 and 396-7. These are repetitive as they give the same explanation. Perhaps discuss the relationship with polarity first, and then the explanation in 392-3 won't be necessary.

We have moved the paragraph containing **lines 392-3** to after the paragraph containing **lines 396-7**. **Line 392-3** still requires the relation of HPLC retention times to polarity as it is not a step in reasoning that is immediately noticeable from Figure 1.A.

Some description of the theoretical hypotheses and reasons for why retention is expected to have a relationship with these properties (through reference to previous retention or other literature) is needed.

We believe that there is suitable general discussion and reference to literature on this topic in **lines 72-74**. This topic is also discussed in the Part I publication (<https://doi.org/10.5194/egusphere-2024-3917>). The chemical properties discussed in section 3.3 are some of the fundamental chemical properties that may determine emergent chemical properties such as H^* , gas or liquid diffusivity, which are commonly discussed in the literature as the most important chemical properties for retention (Stuart and Jacobson 2003,2004; Jost et al. 2017). We feel that adding more discussion to the relevance of these fundamental chemical properties would not adequately service the reader's understanding of their importance to freezing retention, but would distract from the point that H^* may not be as relevant to understanding freezing retention as previously thought.

444-459. The discussion of the relationship with effective Henry's law constant does not adequately address the potential influence of the conditions of freezing and freezing kinetics that are expected to have important impacts on retention coefficients for species with lower effective Henry's constant, as discussed in previous literature. Although the formation of an ice shell as inhibiting expulsion is mentioned at the end here, it appears to be largely an afterthought. The data are consistent with freezing conditions that enhance trapping, increasing retention even for the lowest H^* compounds (which are expected to have high variability in retention dependent on freezing conditions). The assumption that a sigmoidal shape is expected, irrespective of freezing conditions does not do the previous literature on the retention phenomena adequate justice. (See for example Stuart and Jacobson 2003, 2004 (cited in this manuscript) and 2005, doi: 10.1007/s10874-006-0948-0).

We have discussed the influence of physical parameters on freezing retention more thoroughly in the Part I publication. <https://doi.org/10.5194/egusphere-2024-3917>

We agree that the differing physical parameters of this experiment is a significant barrier to its comparison with wind tunnel studies; we present discussion on this at the **end of section 3.3** and in the conclusions in **lines 469-474**. One of the objectives of this study is to establish untargeted complex mixture analysis as a method to measure retention before we make more direct comparisons to the wind tunnel. We have future experiments planned as a separate publication involving wind tunnel experiments. With interest for this discussion, our preliminary data from the tunnel shows similar results to this experiment in the levitator. This gives us reason to believe that the physical differences of these experiments is not the cause for the absence of sigmoidal behavior here.

453-455 (and 469-473). Although this is an appropriate limitation to discuss, it is too narrow. Why focus only on the potential effect of the surface to volume ratio rather than other conditions of freezing, when there are other factors that have been suggested previously as likely important based on theory and modeling?

We have discussed the influence of physical parameters on freezing retention more thoroughly in the Part I publication. <https://doi.org/10.5194/egusphere-2024-3917>

482- 490. Solubility is expected to influence retention but was not studied here and is only discussed as a rationale for why AA don't follow polarity and VP trends. The expected effects of solubility on retention (and its relationship with Henry's law constant) should be discussed more broadly, along with lack or solubility information limiting the findings.

Aqueous solubility is largely dependent on structural properties that this method of untargeted MS is not able to adequately resolve for analysis. The limitation on discussion of solubility stems from the inability to assign high confidence structural information. This is an issue commonly discussed in untargeted complex MS analysis. A comprehensive discussion on this issue would only distract readers from the supported conclusions with speculation that is outside of the current measurement capabilities of this method.

Further, solubility is also already well described in the literature as a piece of the calculation to estimate H^* . We were able to avoid some of the issues with calculating H^* without high confidence structural assignment by accepting the average difference between the H^* for structural isomers as found by Isaacman-Vanwertz and Aumont, 2021 (<https://doi.org/10.5194/acp-21-6541-2021>). This is discussed in **lines 176-180**.

469. 'area'. Typo. I believe you mean surface to 'volume'?

We have incorporated this correction into the manuscript in **line 469**.

490 and 505. ‘insulates’. Wrong word choice. I suggest ‘suggests’.

We have fixed these typos in **lines 492 and 505**.

References:

Szakáll, M., K. Diehl, S. K. Mitra, and S. Borrmann, 2009: A Wind Tunnel Study on the Shape, Oscillation, and Internal Circulation of Large Raindrops with Sizes between 2.5 and 7.5 mm. *J. Atmos. Sci.*, 66, 755–765, <https://doi.org/10.1175/2008JAS2777.1>.

Isaacman-Vanwertz, G. and Aumont, B.: Impact of organic molecular structure on the estimation of atmospherically relevant physicochemical parameters, *Atmos. Chem. Phys.*, 21, 6541–6563, <https://doi.org/10.5194/acp-21-6541-2021>, 2021.