We would like to thank the Editor and R#2 for their thorough and productive comments. We would especially like to thank the Editor for their guidance in this discussion and their advocacy on behalf of R#3's comments. We have addressed the Editor and R#2's comments and suggestions in our responses below. Our revised manuscript is uploaded along with a tracked changes document for the handling editor's consideration. For the responses, the lines, figures, and sections referenced match the numbering in the version of the manuscript most uploaded along with this response without tracked changes. The specific references to the manuscript in our response have been highlighted in yellow for convenience.

Comments made by Referee #2

1. Referee #2 Comment: The response doesn't answer my questions. My concerns remain unsolved.

First, as the authors stated: "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.". Does this mean that the conclusion from this study may not be applied to aerosol particles from different regions where background matrix can be different from Beijing?

Second, If I understand it correctly, the Research articles must include substantial advances and general implications for the scientific understanding of atmospheric chemistry and physics. The authors response: "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.". This is also not a good justification. I strongly suggest the authors either present it as a Measurement Report or at least incorporate these above reasons in their response in the manuscript.

Editor comment: I agree with the referee.

I acknowledge that you do explain nicely potential atmospheric implications based on your observed trends in the conclusion section. However, the potential caveats and limitations of your study due to the unavoidable mixing of samples should be also discussed. Please address it in the Results and Discussion Section and also in the Conclusions.

As requested, we have included more discussion on the purpose and limitations of mixing filter extracts in the experiment. The reviewer asks "Does this mean that the conclusion from this study may not be applied to aerosol particles from different regions where background matrix can be different from Beijing?" They raise a valid concern as there is not enough evidence to assume activity differences due to composition are either negligible or the same elsewhere as in this experiment, but it wouldn't be unreasonable to assume. Afterall, average rainwater DOC is on the order of µM which could be assumed to be dilute enough for activity differences to be negligible. We would also like to distinguish that the concern the reviewer raises is not the result of extract sample mixing, but from unquantified activity differences from sample composition. We have added the following to the Methods (2.1), Results and Discussion section (3.3) and in the Conclusions (4):

> Multiple filters were used to produce the filter extract sample in order to ensure adequate concentration and uniform background signal across multiple measurements. This also controls for potential differences in matrix effects from different filter compositions. 101-103
>
> Additionally, there are chemical dissimilarities in these experiments, as the present experiment is potentially influenced by activity differences from the extract solution's complexity as opposed to the single or few component solutions used by the previously stated studies. 460-463
>
> The use of UHPLC-HRMS has allowed for the study of ambient WSOC retention rather than single component or limited mixture experiments from previous studies. While the influence on retention due to activity differences resulting from matrix effects and solution complexity is still unknown, the experiment in this paper demonstrates the viability of UHPLC-HRMS analysis for ambient WSOC and shows the need for further complex mixture study regarding retention. Future studies on retention within hydrometeors should include complex mixture analysis under the physical conditions most similar to the atmosphere, i.e. wind tunnel experiments, smaller droplets, increased ventilation. As this experiment is a first demonstration of retention within a complex mixture, the applicability of the conclusions here to other locations or samplings with different aerosol compositions—thereby potentially different matrix effects —could be challenged. For example, black carbon particles suspended in a drop could strongly bind organic compounds, preventing their transition into the gas phase during freezing; certain surfactant species could change the surface accommodation, inhibiting exchange; or different amounts of inorganic ions could change the ionic strength of the aqueous phase, altering chemical potential. There is not enough evidence to assume these matrix effects are negligible or the same elsewhere as in this experiment, but the assumption is not unreasonable. Rainwater tends to show negligible matrix effects

> for other properties and analyses (Pang et al., 2017; Sauret-Szczepanski et al., 2006). Average rainwater DOC is on the order of µM which could be assumed to be dilute enough for activity differences to be negligible compared to pure water solutions. However, these are still unsupported assumptions that are required for broad application of these conclusions. 546-560

On the topic of activity differences due to matrix effects and solution non-ideality, we would like to highlight that there is not even enough evidence present to conclusively state whether the retention of single chemical species is different in a complex mixture versus an idealized single component solution, let alone to try to make direct comparisons of different complex mixtures. This is one of the reasons why we find our manuscript valuable to publication. While there is evidence that suggests solution activity differences might be insignificant—specifically, Borchers et. al. 2024 didn't observe any differences in retention between their single component solutions and a mixture of a few nitrophenols; Gautam et. al. 2025 in Part 1 sees little differences between single and binary mixtures—we can't assume that it either is or isn't significant without presenting more evidence. We intend to explore potential activity differences from matrix effects in future publications, possibly in the way the reviewer suggests, but we must first establish that we can measure freezing retention in a complex solution before we can measure riming retention in a complex solution. Our data adds to the body of evidence to potentially answer the question of activity effects and challenges the current understanding of the relationship between H* and freezing retention. Relegating this publication to a Measurement Report because we controlled for variables that would potentially confound the data overlooks the more apparent findings and the reasons why we controlled for the potential differences in background matrix.

2. Referee #2 Comment: As the authors agree the significant barrier to its comparison with wind tunnel studies, then it is not fair to state that the sigmoidal behavior is an overfitting. In the revision, the authors didn't make any adjustment or include the reasoning in the related discussion. If the authors plan to present in a separate publication involving wind tunnel experiments. I would suggest remove the discussion in the comparison with data from the previous wind tunnel studies.

Editor comment: I am still confused how you conclude that N- and S-containing compounds are mostly anthropogenic SOA. There is a huge body of literature that

discuss the formation of such compounds also from biogenic sources (isoprene, terpenes etc). If you are sure that your samples contain organics with N and/or S as heteroatoms, just state it like that in the abstract, e.g. “The findings here also indicate that N- and S- containing organics have enhanced retention.”

I disagree with the referee to remove the wind tunnel data from Figure 10. However, I would like to see a better explanation for the disagreement of low H* species. Why would different physical parameters lead to bad agreement for low H* species but good agreement for high H* species? Were there any other differences in terms of species properties, e.g. water-solubility (which does not necessarily correlate with H*), polarity, functional groups, molecular weight – just to name a few.

The term ‘overfit’ has a negative connotation, as obviously also expressed by the referee who calls it ‘unfair’. Fact is that low retention of low H* species were measured in the previous study. The red line in Figure 10 seems a good fit to this data. However, whether this data – and therefore the fit - is relevant for atmospheric conditions is a different question. I suggest toning down the reference to ‘overfitting’ in your text and phrase it more cautiously.

The N- and S- containing species that we are trying to reference are the ones we discuss in 3.1 (lines 250-290), specifically the nitro and sulphate species evident via MS2 (Fragments with m/z 62 for nitrate or m/z 80 or 90 for sulfate) that we primarily observe along with the amine-nitrates. The species with low H/C and high DBE are aromatic, indicating they are most likely from fossil fuel/biomass burning. The other nitro and sulphate species tend to be NOx and SO2 products that we believe are from local NOx and SO2 emissions. Since sampling occurred in a very urban environment in the winter, we expect the local NOx and SO2 emissions to be anthropogenically dominant. While the isoprene nitrates that the editor references could be purely biogenic, we find this unlikely under these sampling conditions. To clarify, we are considering the products of biogenic precursors that undergo functionalization with anthropogenically sourced N- or S-, namely from NOx and SO2, to be anthropogenically influenced and thus referred to as an anthropogenic SOA component. For example, the terpene-derived organosulfates that the editor discusses are referenced in lines 263-265. For specificity, we have amended line 21:

> The findings here also indicate that N- and S- containing compounds, primarily nitro and sulfate components of secondary organic aerosols (SOA) anthropogenically related to NOx and SO2 chemistry, have enhanced retention likely due to their increased polarity. 21

Additionally, we have further specified 'SOx' as 'SO2' throughout the manuscript. For the purposes of this publication, the distinction between 'SOx' and 'SO2' is insignificant but the reference of "NOx and SO2 chemistry" is more generally used in literature.

As suggested, we have added more explanation for the disagreement of low H* species with wind tunnel studies and to address the question: "Why would different physical parameters lead to bad agreement for low H* species but good agreement for high H* species?" Along with swapping the term 'overfit' with 'fit' as to not imply an undue negative connotation, we have added this discussion to 3.3:

> Physical differences such as higher surface-to-volume ratio, increased ventilation, or longer freezing times may result in lower retentions that may primarily affect species with lower H* as noted by Jost et al. (2017). Small riming droplets have freezing times in the tens of milliseconds while the drops here take approximately 90 seconds to fully freeze. However, the ice shell formation observed by Gautam et al. 2025 in Part 1 is quite fast, in the range of 5 ms. Specifically, these differences either enhance heat and mass transfer which produces a shorter timescale for expulsion or increases freezing time which allows for a wider range of expulsion timescales. Describing this in the framing of Stuart and Jacobson's (2003, 2004) model, species with larger H* are more likely to be unaffected by these differences as the gas-phase mass transport term and the interfacial mass transport term are still dominated by H* despite the decrease in spread droplet height and increase in thermal velocity such that the total expulsion time is still much longer than the freezing time. 477-484

Additionally, we have added more discussion on the differences in terms of species properties of our dataset and the literature we compare against. This is added to 3.3:

> Further, the species studied in the current literature are mostly inorganics which are very different in terms of solubility, polarity, and molecular size compared to organics studied here, i.e. generally more soluble, more polar, and smaller. This could suggest that the organics measured here should have lower retentions than the inorganics in the literature but that is not observed in the data. However, two of the same nitrophenols studied by Borchers et al. (2024) are measured here: 4-nitrophenol and 2,4-dinitrophenol.. It's also likely that 2-nitrobenzoic acid and 4-nitrocatechol or similar analogues are observed within the dataset since other nitroaromatics that cannot be structurally resolved are observed. 2-nitrophenol is also potentially observed, however 2 and 4 nitrophenol are difficult to distinguish from each other as structural isomers and 2-nitrophenol is not easily ionizable in

> this method. However, these assignments cannot be confirmed without better structural information. 463-471

We also edited lines 471-473 to include the omission of formaldehyde from the parametrization by Borchers et al. (2024).

> The literature values include only three species with H* < 10^4 and two of them, pinanediol and formaldehyde, are excluded from the parameterization presented by Borchers et al., (2024) as they were perceived as outliers. 471-473

Comments by Referee #3

General editor comment: Although it is clear that your paper has a companion paper with related experiments, each paper should stand on its own. It cannot be expected that the referee or any reader has to read both papers to understand fundamental findings in the present study. Therefore, please address some of the comments by referee #3 in more detail as suggested below.

Initial referee #3 comment: 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).

Author response: 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.

Editor comment: Given that you highlight this large discrepancy between your current study and those that found a sigmoidal R-H* relationship as one of the main findings, it would seem reasonable to include more discussion on previous literature data such as Stuart and Jacobson 2005 and possibly others.

We have included more discussion on how the physical factors may account for the nonsigmodial behavior in terms of Stuart and Jacobson (2003, 2004) model. Please consider the previous comment.

Initial referee #3 comment: 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?

Author response: 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

Editor comment: A brief discussion should be added also in this paper. It does not have to be as detailed in Part I.

We have included more discussion on how the physical factors may account for the nonsigmodial behavior in terms of Stuart and Jacobson (2003, 2004) model. Please consider the previous comment.

Initial referee #3 comment: 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.

Author response: 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.

Editor comment: I appreciate that you explain here that you explain here why solubility is not taken into account as a parameter. However, this information should be also mentioned in the manuscript text in a couple of sentences. I don't think this would be distractive.

Water solubility and Henry's law constants do not necessarily correlate. The former describes how much of a substance can be dissolved in water and above which limit it forms a solid or supersaturated

solution ; the latter describes how a compound partitions between the gas and aqueous phases. Thus it describes two different thermodynamic concepts.

There are very distinct differences. For example, small alcohols are fully miscible in water but have very low H*; in contrary, some salts are highly water soluble, others not – but all of them basically have infinite Henry's law constants.

The limitation and implication of analysis on solubility is already discussed briefly in terms of AA in the above discussed passage. As suggested, we have broadened this discussion with a few more sentences on solubility's influence on H* and the limitations from the current method:

> Indeed, solubility as a property relevant to retention is applicable to all compounds, specifically in how aqueous solubility is related to H*. While aqueous solubility is a piece of the typical bond method calculation to estimate H*, it does not necessarily correlate with true H*. Further analysis on solubility as a factor for retention would be valuable but the limitations on high confidence structural assignment in this method prevent its thorough investigation. Structure assignment could also

> elucidate other retention relevant properties such as hydrogen bonding potential or even acid/base effects. 515-521

l. 13/14: The structure of this sentence does not seem right; ‘incurs’ might not be the right verb here. Better use ‘causes’ or similar.

We have swapped the use of “incurs” to “causes”. 13

l. 15: The number and variety of compounds that can form new particles is very limited. Why are so specific here? Do you have evidence that indeed new particle formation occurs in the upper troposphere by compounds released from freezing droplets?

Just the fact that compounds can be released upon vertical transport seems a sufficient motivation to study retention coefficient. These compounds may then take part in any chemical reactions.

This study of retention transport was specifically motivated as an explanation for previously unexplained NPF. Specifically, Williamson et al. (2019) and Bardakov et al. (2021), which we discuss in the introduction in lines 42-55, propose the underestimation of available organics as an explanation for NPF in the upper troposphere from convective outflows.

We feel the specific motivation is sufficient, but we have generalized the phrase from “available for new particle formation” to “available for atmospheric processes int the upper troposphere such as new particle formation or ozone formation”. 15

l. 25: differs from… (not ‘with’)

We have swapped “with” with “from”. 25

l. 26: I am missing in the abstract a concluding sentence that summarizes why the current study is more reliable/refined/better/… than the previous literature studies.

We have added a line at the end of the abstract to summarize the improvements of this study to the literature.

> This study greatly expands on the available experimental measurements of retention by investigating hundreds of compounds in complex chemical conditions more similar to the atmosphere than the previous literature studies. 27-29

l. 34: SOA formation is not necessarily part of new particle formation. Compounds with sufficiently low vapor pressure may condense on any particle, whether primary or secondary. Also the initial particle may be inorganic (e.g. by new particle formation due to sulfuric acid) or organic.

We have replaced the reference to new particle formation from this line with a reference to inorganic contributions.

> These effects are controlled by factors such as their optical properties, size, and the hygroscopicity (Dusek et al., 2006; Sun et al., 2021), which can change based on the proportions of primary organic aerosols (POA)—directly emitted aerosols—and secondary organic aerosols (SOA)—aerosols formed from the oxidation products of volatile organic compounds (VOC)—as well as inorganic constituents (Hallquist et al., 2009; Liu et al., 2021; Riva et al., 2019). 33-39

l. 49 – 51: I am not convinced that uncertainties in gas transport are the main factor of uncertainty of NPF in the upper troposphere. Isn't it much more the uncertainties in NPF precursors, i.e. which compounds nucleate new particles at what rate?

We have edited this line to be more specific to NPF precursors.

> If this NPF is the result of an overlooked mechanism of organic matter transport, it is then critical to elucidate this mechanism for NPF precursors so to constrain uncertainty around the influence of high altitude NPF from convective outflows (Bardakov et al., 2021). 53-55

l. 56: 'incurred' seems wrong here. Do you mean 'occurred'?

We believe 'incurred' is used properly here, but we have swapped its use to 'caused'. 60

l. 74: What is the difference between 'liquid water content' and 'droplet size' here? Isn't the liquid water content simply the water volume and thus proportional to size?

Unless you mean something different, I suggest removing liquid water content here.

The misunderstanding here is that LWC is a property of an individual drop and not of the environment the drop is in. We have struck the phrase "of the droplet" from this line to avoid misunderstanding. (line 78) This phrasing also mistakenly implies ventilation is a property of a single drop.

Liquid water content is more similar to humidity in that it's a mass concentration of water in a volume of air. Droplet size is often related because of growth or evaporation effects.

l. 82: 'cloud size range' seems odd here. Do you mean 'droplet size range'?

We mean the size ranges typical for droplets within natural clouds. We have rephrased the line as:

> Additionally, current studies have only examined retention for cloud droplet sizes rather than raindrop sizes. 86

l. 251: Do nitrophenols from other sources (e.g. industrial emissions) have different H/C ratios?

I don't believe so but this would be interesting to consider. The H/C ratio is derived from condensed nature of aromatics so all nitrophenols regardless of origin should be similar in terms of H/C ratio.

l. 253/254: Are the compounds specific to automobile and shipping emission or could it be also other organosulfates?

The alkylorganosulfates compounds referenced are specific to high sulfur fuel combustion, which is almost exclusive to automobiles and ship emissions. To be specific, saturated organosulfates C>5 are not of biogenic origin, but rather shipping, traffic, or coal combustion. This is discussed in the references presented: Blair et al., 2017; Qi et al., 2021.

If the editor is insinuating the other organosulfates to be biogenic terpene products, we do discuss those in the lines following.

l. 256: Sulfonic acid (oxidation state of S = +4) is not sulfate (oxidation state +6)

We have added the phrase "in addition to compounds such as camphorsulfonic acid" as to not mistakenly imply that camphorsulfonic acids is a sulfate. 263

l. 257: The study by Iinuma et al was conducted at a forested, not urban, site. Given that terpenes are biogenic compounds, I do not think that camphorsulfonic acid is only formed under urban conditions.

We have amended the line as:

> …such as camphorsulfonic acid (C10H16O4S, 232 m/z, 9.4 min, L2), which also demonstrate secondary processing with sulfate aerosols (Iinuma et al., 2007; Surratt et al., 2007). 263-264

l. 266/7 and following: Just providing the sum formula of the compounds is rather confusing since these formula could also represent compounds with functional groups other than amines (cf e.g., https://en.wikipedia.org/wiki/C9H11NO2#) Please

either write the formulae such that the molecular structure is clear and/or add the compound names.

Our method is limited in which structures it can confidently assign. We can assign the sum formula with high confidence and that these compounds are amines based on their ionization mode, H/C ratio, O/C ratio, and any available MS2 spectra. However, a full structure cannot be assigned with high confidence without standards for each compound of interest. We don't want to imply more structural information than we have by giving these compounds more description than the sum formula. We believe that stating the sum formula and class of compound is a more precise description of the present information than presenting a misleading structure candidate. This is consistent with our usage of the convention of Schymanski et al. (2014) to communicate structural confidence.

l. 277: One of the main oxidation products of isoprene include methyl vinyl ketone (among others), with an H/C ratio of 1.5. Thus, I don't think that the H/C range is a valid indicator of biogenic vs anthropogenic. If I am wrong, please add more recent references demonstrating it.

The editor is correct in stating that MVK has an H/C ratio of 1.5. However, the distinction of a CHO species with H/C ratio above or below 1.5 is not being used as an indicator of biogenic vs anthropogenic origin in this instance but compound classification of aromatics versus aliphatic acids. In the next line, we are stating that we see few species within a specific range of H/C that the compound classes of humics and lignin that typically have. These classes are then associated with biogenic origin.

We have added a reference to Ma et al. (2022) (Line 287) which more thoroughly investigates the markers of wintertime Beijing aerosol through untargeted MS analysis. This section is primarily intended to describe that our sample set is typical of Beijing winter aerosols.

Tables 2 and 3: These tables could be moved to the supplement as they are not discussed in the text, but oly referred to in a very brief sentence (cf also comment by Referee 3). Their main information is included in Figure 3 . Readers who are interested in the data can find them in supplement.

We have moved tables 2 and 3 to the supplement and adjusted line 328 accordingly.

l. 333/34: CHNOS represents more of the heaviest species in the sample set while CHN is entirely AA.

This sentence is quite cryptic. Please reword.

We have reworded this line for clarity as:

> These CHNOS compounds have some of the highest molecular masses in the dataset, evident in Figure 1. The CHN species present are all AA. 340-342

l. 345: The paper by Daito et al does not refer to the atmosphere. The experiments were performed under very different conditions, not relevant to atmospheric ones. Unless you can add a relevant reference here that demonstrates that aqueous phase reactions in the atmosphere lead to NOx removal, I suggest removing this part of the sentence.

We have reformulated the sentence with more relevant references:

> It is known that NOx participates in the reversible and irreversible uptake of isoprene to aerosol liquid water and can further react with isoprene to produce a substantial number of organonitrates. (El-Sayed et al., 2018; Tsiligiannis et al., 2022). While it's also known that organic nitrogen represents an important fraction of WSOC (Saxena and Hildemann, 1996; Zhang et al., 2002), this data may also indicate that nitrogen chemistry on CHO species enhances their retention in hydrometeors. 351-356

l. 419: The paper by Weschler and Nazaroff is about semivolatile organics only and in indoor environments. During the last decade there have been many papers on the volatility ranges of SOA species in the atmosphere. It would seem more appropriate to cite one of them.

We have updated the reference to two more recent publications on LVOCs and IVOCs. (Li et al., 2023; Manavi and Pandis, 2024). 427

Conclusions: Define AA and VP here once more for the readers who only read the conclusion section.

We have made this adjustment in lines 497 and 509.

l. 493: 'nitrogen and sulfur inclusion' is not a commonly used term. I think what you mean is 'functional groups containing sulfur or nitrogen'

We have incorporated this into the manuscript in line 526.

l. 494/5: cf my comment on the abstract

We have adjusted this sentence for clarity and specificity in agreement with the previous.

> Overall, the high retentions of the nitro and sulfate species typically anthropogenically related to NOx and SO2 chemistry, indicate that NO_x and SO_x

> chemistry may enhance the retention of these SOA species, reducing their likelihood of reaching the upper atmosphere. 527-529

l. 497: cf my comment about this reference above

We have reformulated the sentence with more relevant references as previously done:

> Further on this, other studies have demonstrated that NOx participates in the reversible and irreversible uptake of isoprene to aerosol liquid water and can further react with isoprene to produce a substantial number of organonitrates. (El-Sayed et al., 2018; Tsiligiannis et al., 2022). 529-532

l. 501 – 503: Atmospheric chemical processing generally tends to oxidatively degrade large nonpolar species into more water soluble, less volatile species (Iavorivska et al., 2016)

This sentence is not quite correct. Chemical processing does not necessarily lead to degradation of organics but can also lead to functionalization. For this you could cite any atmospheric chemistry textbook rather than the paper by Iavorivska et al that is about deposition.

We have edited the sentence to include functionalization and updated the reference to "Atmospheric Chemistry and Physics: From Air Pollution to Climate Change" by John H. Seinfeld and Spyros N. Pandis.

> Atmospheric chemical processing generally tends to functionalize or oxidatively degrade large nonpolar species into more water-soluble, less volatile species (John H. Seinfeld and Spyros N. Pandis, 2019). 537-539

l. 508/9: Why do you limit your discussion here to SOA precursors? By far not all organics form SOA. Just the fact that less oxidized (or 'fresher') organics are less likely to be retained and vertically transported than more aged organics is an interesting result. Whether they eventually form SOA is not relevant.

We have broadened the discussion here by swapping the instances of 'SOA precursors' to simply 'organics'.

> This indicates that many freshly oxidized organics may have a lower potential to be retained than aged organics and may generally suggest that freshly oxidized organics are more likely to reach the upper atmosphere than primary organics or aged organics. 543-545

l. 515 – 520: I got confused by this text (and obviously also referee 2). What do you want to say here? Would you expect that during summer the WSOC composition is

completely different and therefore your results are irrelevant? What is known about WSOC differences between summer and winter?

This bit of discussion was added in response to a concern raised by Reviewer #1. They noted that the conditions required for the retention transport that we describe occur more frequently in the summer rather than in winter, which is when our samples were taken. Specifically, the convective systems that would facilitate the transport of surface emissions to altitudes where ice cloud formation occurs are not typical for the winter. That's not to limit the relevance of our results, but to highlight that our results may not be entirely representative of all the relevant species that participate in this process.

We have expanded on this discussion with the following:

> Additionally, broader application of these conclusions should consider the differences in relevant WSOC composition regarding seasons. This sample set is representative of winter aerosols, which show a high contribution from nitroaromatic compounds as well as a lower degree of oxidation and a lower proportion of organosulfates in comparison to the summer (Ma et al., 2022). 562-564

l. 531ff: Can you explain better what you mean here? What should be included in a transport model? Usually retention coefficients are included for individual species or species groups, independent of their mass distribution. Retention is usually just described as a mass fraction that remains in the ice phase. Why should this depend on species abundancy?

Are you saying that the retention coefficients measured in your study are only valid for this particular composition? If so, why? It would imply that the presence of all compounds in a sample affects the retention of an individual compound.

The discussion here was aimed at qualifying the application of the parameterizations presented in Table S1 and S2. The parameterizations simply provide the distribution for all measured species without regard to which species are more abundant. It most likely is best practice to implement the retention coefficients as individual species or species groups, but we wanted to make that distinction for someone looking to estimate the retention of a random organic by applying the frequency distribution of this dataset.

To clarify this, we have swapped out the instances of 'measurements' and 'data' with more specific phrases:

> These parameterizations of retention also present the distribution of retention coefficients for the variety of species present and not necessarily the mass

distribution of species potentially present in the atmosphere. Corrections for species abundancy must first be made in order to apply the frequency distribution of the retention coefficients here to organic transport models. 580-584

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