We are grateful for the editor's decision as well as taking the time to check our revised manuscript thoroughly. We have carefully addressed the editor's valuable comments and suggestions in the responses provided below. **Red colored** text indicating **editor's comments and suggestions**, and **black font** indicating **our responses** to them. ***Rewritten and newly added texts*** in the manuscript are provided below in ***italics*** for convenience. Line numbers mentioned here correspond to the revised manuscript. A revised version of the manuscript will be uploaded for the handling editor's consideration. **"Main editor comments: no.2, 1st part and "Minor/Technical comments: l. 209-215", are highlighted** for ease of navigation.

Main editor comments: (Line numbers refer to the manuscript version with track-change).

Editor: 1) Title: The referee asked 'Retention of what'?

I think the changed title still doesn't answer the question. Wouldn't it be clearer to say

Retention of Organic and Inorganic Trace Gases during Freezing of Rain Drops: Part 1 Investigation of Single and Binary Mixtures.

Or even

Retention of nitric, formic and acetic acids and nitrophenol....

I am aware that you have a 'Part II' paper in review as well. However, you may consider changing its title accordingly.

Response:

Considering the suggestions above, the title has been changed for better clarity.

"Retention During Freezing of Raindrops, Part I: Investigation of Single and Binary Mixtures of Nitric, Formic and Acetic Acids and 2-Nitrophenol"

Editor: 2) I am confused about your discussion on the effective Henry's constants, e.g. in sections 3.1 and 3.4 but also in the introduction where you refer to previous studies. You say that in previous studies, compounds showed a dependence on pH due to increasing solubility (KH*) with increasing pH. I argue that some of the observed pH dependencies are not a function of solubility or KH*. For example, within the pH range of your experiments ($3 < pH < 6$), KH* for nitrophenol does not change (see figure below – note that, for simplicity, I used values at 25C and expressed KHeff in M/atm; however, the trends are likely similar at lower T, and are independent of the unit). Therefore, if retention were a function of solubility, no change in R should be expected. The same applies for acetic acid at pH < ~5.

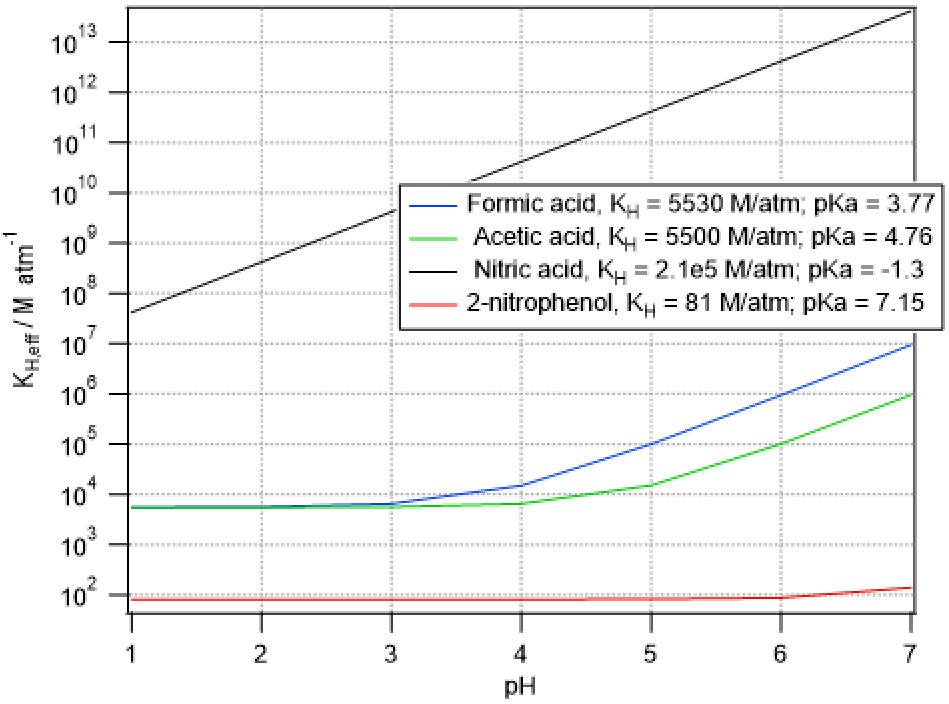


Figure A. Relationship between effective Henry's constant and pH. (We labelled this figured for the ease of reference)

If I understand correctly, you used your measured R values and KH* values to derive the empirical coefficients a and b in equation 4. Which R values did you use as input to the equation? In your Fig 2, you show that there is a statistically significant difference for R(nitrophenol) as a function of pH – while KH*is identical. It would be useful if you commented on this and maybe even include a figure as above (as a supplement) to add to the discussion.

Response:

We are greatly thankful for your insightful comment and the plot (Fig. A). We have included a similar plot in the supplement as Figure S1. We are splitting our responses into two parts: **1st part** - regarding the 'observed pH dependencies are not a function of solubility or KH*' and **2nd part** - regarding equation 4, (a and b parameters).

We also found the comments regarding the **1st part** here, concerning KH* and pH, and in **"Minor/Technical comments: l. 209-215"**, concerning pH dependencies to be closely related to each other. Hence, our responses for both these similar sections are collectively stated here, to avoid repetition.

1st part:

(and "Minor/Technical comments: l. 209-215:")

We agree to the statement "some of the observed pH dependencies are not a function of solubility or KH*". From fig. A, one should ideally observe a pH dependence for formic and acetic acid and none for 2-nitrophenol for our measured pH range. Our ignorance in performing a proper statistical analysis for pH dependencies, led to misinterpretation of our data.

We did a careful investigation of our data and performed linear regression test (95% confidence interval) using IBM SPSS Statistics-Version23 for the investigated single components and their dependence on pH values. p- value lower than 0.05 indicates dependence of retention coefficients on pH. The results for single component substances are as follows:

Acetic acid:

The linear regression tests reveal a significant statistical dependence of the retention of acetic acid on pH, with p = 0.047. This result contradicts what we had previously assumed for dependence of acetic acid on pH (owing to large standard deviations). The linear regression test results are in agreement with "Minor/Technical comments: l. 209-215: 'I see it the opposite way based on the figure, i.e. that there is barely any dependence of R on pH for nitrophenol (at least at pH < 5); however, there is a steady increase of R with pH for acetic acid.' Acetic acid is not completely retained at pH 4, so an increase in retention can be seen at higher pH which is also supported by Fig. A.

Text has been added/changed in L206 as:

"Linear regression test (SPSS V23) reveals a significant statistical dependence of the retention of acetic acid (green marker) on pH, with p = 0.047. Acetic acid was not completely retained at pH 4.2 (R = 0.88), and an increase in retention was seen at higher pH. With increasing pH, the H∗ also increases for acetic acid, (see Fig. S1). The retention coefficients for acetic acid were 0.81, 0.88, and 1.05 for pH values of 3.1, 4.2, and 7.0, respectively, while their corresponding standard deviations were 0.18, 0.12, and 0.2."

Formic acid:

Linear regression test for formic acid gave a p-value of 0.182, indicating no significant dependence for pH, also seen in Fig 2 in the manuscript. Formic acid is already completely retained (R =1) at pH 4. As such, any increase in pH would not lead to an enhancement of the retention, even though KH* for formic acid varies in a similar fashion to acetic acid (Fig A).

Text has been added/changed in L211 as:

*"Formic acid (blue marker) did not show any dependency on pH (p = 0.182). Formic acid is already completely retained at pH 4.1 (R= 1.01), and as such, any increase in pH would not lead to an enhancement of the retention, even though H*for formic acid varies in a similar fashion to acetic acid (Fig. S1)"*

2-nitrophenol:

Interestingly, 2-nitrophenol (with p = 0.005) also showed statistically significant dependence of retention on pH, for our measured pH range from 3 to 6. This result for 2-nitrophenol is contradictory to the expected form of dependence of KH* on pH seen in Fig. A. Here, a probable explanation is that the 2-nitrophenol is more dissociated at pH 6 than at pH 3 and 4. We calculated the fraction of deprotonated to protonated ions at pH 3, 4 and 6 for 2-nitrophenol. This ratio was found to be 7×10^{-5}, 7×10^{-4}, 7×10^{-2}, at pH 3, 4 and 6 respectively. This means that at pH 6, about 7% of 2-nitrophenol is present in deprotonated form. During the freezing process, deprotonated molecules must undergo protonation to achieve neutrality before they can be expelled from the drop. At pH 6, a higher proportion of molecules remain confined within the drop due to the requirement for proton recombination prior to volatilization and subsequent expulsion. This pH dependence for 2-nitrophenol is also in agreement with Borchers et al. (2024), where they measured retention coefficients of α-pinene oxidation products and nitro-aromatic compounds during riming for cloud droplets.

Text has been added/changed in L214 as:

"2-nitrophenol (red marker) showed statistically significant dependence of retention on pH (p = 0.005), for our measured pH range. The retention coefficients of 2-nitrophenol at pHs of 3.2 and 4.4 and 6 were 0.90, 0.90 and 1.05, respectively, and their corresponding standard deviations were 0.08, 0.05 and 0.11. This result for 2-nitrophenol is contradictory to the expected form of dependence of $H*$ on pH, as in Fig. S1. 2-nitrophenol is more dissociated at pH 6 than at pH 3.2 and 4.4. The fraction of deprotonated to protonated ions at pH 3.2, 4.4 and 6 for 2-nitrophenol was found to be $7x10^{-5}$, $7x10^{-4}$ and $7x10^{-2}$, respectively. This means that at pH 6, about 7% of 2-nitrophenol is present in deprotonated form. During the freezing process, deprotonated molecules must undergo protonation to achieve neutrality before they can be expelled from the drop. At pH 6, a higher proportion of molecules remain confined within the drop due to the requirement for proton recombination prior to volatilization and their subsequent expulsion. This pH dependence for 2-nitrophenol is also in agreement with Borchers et al. (2024), for riming retention of cloud droplets"

Summary:

Given the overall high retention for formic, acetic acid and 2-nitrophenol, the dependency on pH for raindrops might not be critical factor, as compared to cloud droplets. The ice shell formation remains the major contributing factor for high retention in raindrops.

2nd part:

We did not derive the parameters a and b mentioned in equation 4. Rather, the values are taken from Borchers et al. (2024). They used the retention values from their study, along with previously measured retention coefficients from von Blohn et al. (2011,2013) and Jost et al. (2017) also involving cloud droplets – and updated the parameters a and b in light of their findings. We plotted this updated fit alongside our present data for retention of raindrops for comparison purposes – and to show that the dependency of retention on solubility and dissociation (i.e. on H*) do not entirely hold true for mm sized drops.

Text has been changed in L248 to better clarification as:

"The relation between effective Henry's law coefficient and retention coefficient for cloud droplets i.e., retention-riming, was modeled by the following equation:"

In L251 as:

"Values a and b were taken from Borchers et al. (2024)."

And in L255 as:

"Equation 4 was plotted in Fig. 4 against our current data for comparing the dependency of R on H∗, for µm sized droplets and mm sized drops."

Editor: 3) There are several studies that revealed that the gas-aqueous partitioning of nitrophenols in cloud droplets may not adhere to their Henry's law constants, e.g.

Lüttke et al. Phenols and Nitrated Phenols in Clouds at Mount Brocken, Intern. J. Environ. Anal. Chem..Vol. 74(1-4). pp. 69-89 Lüttke et al. Phase partitioning of phenol and nitrophenols in clouds, Atmos. Environm., 1997, 2649-55.

I wonder if the behavior of nitrophenol in your study could be partially explained by this.

Response:

Thank you for the comment and the references.

We did go through the suggested studies - Lüttke et al.(1997 and 1999).

Lüttke et al. (1997) showed that liquid-gas partitioning coefficient for 2-nitrophenol is about 6 times higher for their observations. A possible explanation was due to the adsorption of 2-nitrophenol on the surface of the droplets. They could not give satisfactory explanation for this increase. In Lüttke et al. (1999), they found that 2-nitrophenol in liquid phase can be approximately described by H*. In both these studies, they refer to measured H* values for 2-nitrophenol from Tremp et al. (1993) and Schwarzenbach et al. (1988). More recent measurements (eg. Guo and Brimblecombe, (2007) in Sanders (2023)) for 2-nitrophenol show 2-fold higher H* compared to the above mentioned earlier measurements cited in Lüttke et al. (1997 and 1999) studies. Also, Lüttke et al. (1997 and 1999) measured droplets transported over time at mountain ranges. These carried over droplets could have other dissolved substances as well, which also might have had an influence on their results.

Retention of 2-nitrophenol has also been studied for riming retention, which follows the sigmoidal dependence on H* in Borchers et. al. (2024). They reported a low retention coefficient of 0.12 for 2-nitrophenol at pH 4. However, in our case, we found the longer solute expulsion timescale compared to the ice shell formation time leads to the high retention values observed for 2-nitrophenol, and perhaps not the influence of H*.

Minor/Technical comments

Please carefully proofread the paper. In particular pay attention to the correct use of articles.

Editor: a) I list a few places below where ‘the’ or ‘a’ is missing, e.g.

l. 20: from the boundary layer

l. 36: in the context

l. 331: could have the potential

l. 333: cracking of the ice shell

l. 337: where the fraction of liquid freezes and the majority …

Editor: b) Also please pay attention to the consistency of singular/plural forms of subject and verb, e.g.

Editor: l. 61: Freezing of raindrops is...

Editor: Table 2: ...temperature was

Editor: l. 323: ...rates ... imply

Response:

Thank you for pointing out these small yet very important grammatical errors. Changes have been made accordingly in the above mentioned points.

Editor: Abstract:l. 1/ 2: You may want to consider improving the first sentence (in particular since it is the first sentence), *The interaction with freezing processes and* *vertical transport of trace gases into the upper atmosphere during deep convection is critical to understanding the distribution of aerosol precursors and their climate effects.*

1) 'Interactions with..' does not seem right here

2) Processes do not really 'interact' – they may be coupled or influence/affect each other.

If I understand correctly, you want to say "Freezing processes affect the vertical transport of trace gases into the upper troposphere…

Response:

Thank you for pointing it out. Text has been changed in L1 as:

"The influence of freezing processes and vertical transport of trace gases…"

Editor: l. 10: *"Thus, for rain sized drops almost everything is fully retained during the freezing process, even for species with low effective Henry's law constants."*

This sentence sounds quite colloquial. Given that you define 'retention coefficient', it may be clearer or more precise to say that the retention coefficients for all single compounds and mixtures were near 1 (or give a range)

Can you specify 'low effective Henry's law constant'?

Response:

Text has been changed in L9 as:

"Thus, for rain sized drops almost everything is fully retained during the freezing process i.e., retention coefficients close to 1, even for species with low effective Henry's law constants, $H^* < 10^{-4}$."

Editor:l. 27: 'evident' seems redundant here

Response: The word 'evident' has been removed in L27

Editor:l. 30: Here you use 'drop' in the context of clouds – given the referee comment and your response, shouldn't it be 'droplet'?

Response:

Here we introduced the definition of retention coefficient in a general sense. We do see the fact that it might lead to confusion. Text has been changed in L28-31 as:

“Trace gases dissolved in these droplets could be either retained, revolatized, or scavenged during the freezing process (Pruppacher and Klett, 2010). The fraction of chemical species remaining inside the frozen hydrometeor, compared to their initial concentration in liquid phase before freezing, results in the so-called retention coefficient.”

Editor:l. 32, 37 (and maybe other places in the manuscript): For better readability, please move the references to the end of the sentence.

Response:

Thank you for pointing it out. References have been moved to the end of the sentences in L32, 37 and other places as well.

Editor:l. 43: ‘Additionally’ implies that H* is neither a chemical nor physical property as they were already mentioned in the previous sentence. Thus, ‘additionally’ seems redundant here.

Response: The word ‘additionally’ has been removed in L43.

Editor:l. 55: ‘A significant difference from a physical perspective in terms of retention of trace gases for cloud droplets and rain drops would be the initiation and pathway of freezing’

- Why do you use subjunctive (‘would’)? If it is a well-known fact, ‘is’ is appropriate. (Please check the full manuscript for use of ‘would’ and decide whether the use of indicative (‘is’) is appropriate.)

- ‘initiation and pathway of freezing’ - is usually referred to as ‘freezing mechanism’.

Response:

Thank you for pointing it out. Text has been changed in L54 as:

“A significant difference from a physical perspective in terms of retention of trace gases for cloud droplets and rain drops is the freezing mechanism.”

As per suggestion, text has been changed/modified for instances of ‘would’, changes can be seen in the track-changes document.

Editor:l. 57: ‘was implemented’ sounds odd. You may implement something in a model but this is certainly not meant here... isn’t something like ‘was the main mechanism’ or ‘took place’ more appropriate?

Response:

Text has been changed in L56 as:

"For riming experiments involving cloud droplets freezing is initiated upon contact with a frozen substrate, whereas, for rain drops investigated in this present study, immersion freezing was the main mechanism."

Editor:l. 67: It may be useful to add the H* values here already, together with the pH value.

Response:

Text has been added in L65 as:

*"To visualize our experimental outlook, we selected four chemical substances namely: 2-nitrophenol, acetic acid, formic acid, and nitric acid, with increasing H*values of* 3.50×10^{3}, 1.28×10^{5}, 8.31×10^{5} *and* 7.56×10^{11}*, respectively, at 0 ◦C and pH about 4, for all."*

Editor:l. 69: what do you mean by '...values for riming with cloud droplet sizes' – is it simply 'in riming cloud droplets'?y

Response:

Text has been changed in L68 as:

"These substances are commonly found in the atmosphere and their previously measured retention coefficient values in riming cloud droplets lie between 0 to 1 and scale with H."*

Editor:l. 104: - ppm and ppb are mixing rations, not concentrations.

- Please specify that you mean 'ppb to ppm (on mass basis)' to make clear that you mean 1 g in 10^9 g or 10^6 g, i.e. 1 ug/L or 1 mg /L (assuming that water density = 1 g/cm3)

This avoids confusion since gas phase mixing ratios of trace gases are commonly given in ppb whereas e.g 50 ppb ozone means '50 molecules out of 10^9 molecules'

Response:

Thank you for pointing it out. Text has been changed in L102 as:

"Typical mixing ratio of dissolved gases in the atmosphere lies in the range of ppb to tens of ppm (on mass basis)"

Editor:l. 111: ‘least’ should be ‘lowest’

Response:

Text has been changed in L110 as:

“- which has the lowest molar mass among the investigated species – “

Editor:l. 120: ‘benzoic’ misspelled

Response: Thank you for pointing it out. Spelling correction has been made for “*2-nitrobenzoic acid”* in L119.

Editor:l. 141: You used D already for drop diameter (abstract). I suggest changing it there and simply spell out ‘drop diameter: 2 mm).’

Response: Thank you for your suggestion. Changes have been made in L3 as:

“...for freely levitating rain drops (drop diameter: 2 mm) using an...”

Editor:l. 164/166: Even though Referee #1 did not specifically comment on the text in these lines, ‘average freezing temperature’ should be also replaced here by ‘median freezing temperature’.

Response: Thank you for pointing it out. Changes have been made in L162 and L164.

average freezing temperature replaced with “*median freezing temperature.”*

Editor:l. 169: Please clarify this sentence: “ The 50% frozen fraction at-23∘C was found to be −6.9±1.1∘C.”

Response:

“50% frozen fraction” has been used to synonymously refer to the median freezing temperature, as stated in L161. However, to avoid any further confusion the term “50% frozen fraction” has been replaced with median freezing temperature.

Changes have been made in L161 as:

“From the temperature profile obtained for experiments conducted at -15 ^{0}C cold room temperature and 0.2 g/L AgI, the median drop freezing temperature was found to be −3.9 ± 0.3 ^{0}C, under these experimental conditions (Fig. A2).”

and in L166 as:

“The median drop freezing temperature for -23 ^{0}C cold room temperature was found to be −6.9 ± 1.1 ^{0}C.”

Editor:l. 171: ‘sized’ can be omitted here and also in the remainder of the manuscript for similar instances.

Response: Thank you for your suggestion. The word ‘sized’ has been removed in L168 and 3 other instances with similar context.

Editor:Table 2: Clarify in the caption whether the R values are averaged over all pH values or only apply to a specific pH.

Response:

Table 2 caption has been changed as:

“Retention coefficients at drop freezing temperature of −3.9 ± 0.3 ^{0}C and pH values about 4 for all the investigated substances. The corresponding walk-in cold room temperature (ambient temperature) was −15 ± 1 ^{0}C.”

Editor:l. 182: “Brand (2014) studied the retention of large drops (2.67 mm and 7.25 mm spherical equivalent diameter)” – please clarify what Brand investigated. It should be the retention of gases (organic acids? All the same compounds as you used in the present study?) in large drops.

Response:

Brand studied the retention of formic, acetic, oxalic and malonic acids. Text has been added in L179 as:

“Brand (2014) studied the retention of formic, acetic, oxalic and malonic acids – for large drops (2.67 mm and 7.25 mm spherical equivalent diameter) by freezing them on a Teflon coated pallet – also reported high retention coefficients (close to 1)”

Editor:l. 185: ‘with which …was realized’ can replaced ‘representing’

Response:

Text has been changed in L182 as:

“However, in our study contact-free immersion freezing was employed, representing a more realistic scenario to initiate freezing as compared to Brand (2014).”

Editor:l. 193” replace ‘least’ by ‘lowest’

Text has been changed in L190 as:

“...having the lowest H^* among the investigated substance...”

Editor:l. 209-215: What is the main message here?

First you say that *“Acetic acid (green marker) and formic acid (blue marker) did not show any apparent dependency on pH”*

Then you say that *“The retention coefficients for acetic acid were 0.81, 0.88, and 1.05 for pH values of 3.1, 4.2, and 7.0, respectively, while their corresponding standard deviations were 0.18, 0.12, and 0.2”* – doesn’t this trend show a dependence? I understand your argument that the standard deviations are larger than the differences between the mean values – however, yet, the figure shows a clear trend and an average R at pH = 7 that is about 25% higher than that at pH = 3.1.

“From Fig. 2a, one can infer a slight dependency on pH for 2-nitrophenol, and almost none for acetic acid and formic acid.”

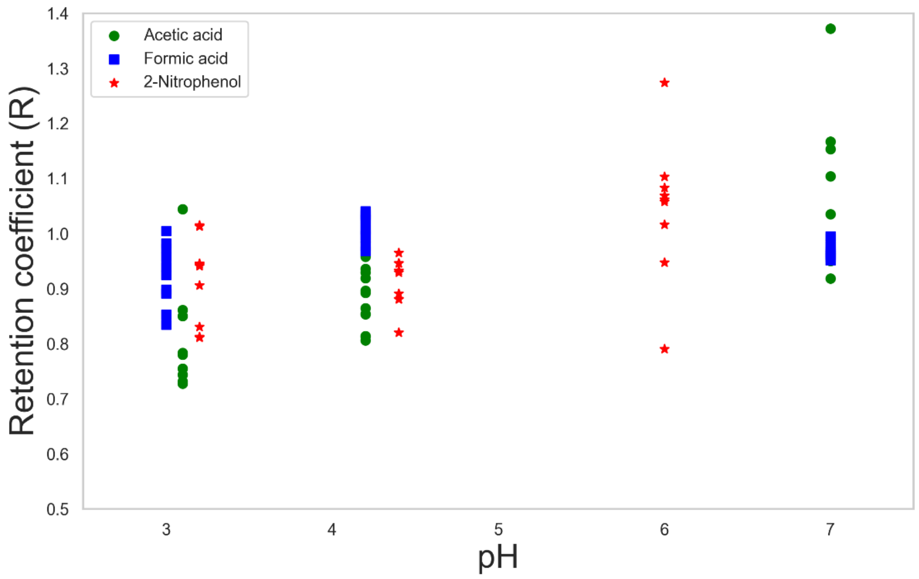
I see it the opposite way based on the figure, i.e. that there is barely any dependence of R on pH for nitrophenol (at least at pH < 5); however, there is a steady increase of R with pH for acetic acid.

I understand your argument that your conclusions are based on the results of 11 experiments. Why don’t you show these values rather than just the averages ± standard deviation that (falsely?) imply a trend and therefore contradict your text?

Response: We are thankful for your insightful comment.

In our response to “Main editor comments: **no.2, 1st part**, we have addressed this concern extensively and added new text to the manuscript accordingly for better clarity. Kindly refer to our response for the highlighted section.

Below we provide the plot showing our measured retention coefficients at different pH values for the single components.



The overlap of the data points doesn't seem very neat and informative in the new plot. As such, we would prefer to include the original plot for Fig 2 in the manuscript, with the averages ± standard deviation, provided there aren't any further objections. We have provided this figure in the supplement as Figure S2, for reference.

Editor:l. 219 - 224: It is difficult to understand what you are saying here.

"pH of the solutions were altered by adding HCl and NaOH, which could also interact with the investigated substances and dissociate them into their ionic form"

Isn't this idea of pH adjustment that you change the proportions of dissociated vs undissociated forms? I suggest omitting this sentence as the second part is confusing (if not even wrong as adding NaOH does not lead to dissociation but association of H+ and carboxylates), and the first part was already mentioned in Section 2.

" In this case the overall concentration of the investigated substances could be lowered." Which concentration is lowered under what conditions? When acids dissociate (i.e. at enhanced pH) the total aqueous phase concentration (acid + anion) actually increases.

Or are you saying that the solubility of the solutes is expected to decrease in the presence of additional solutes such as HCl and NaOH due to salting-out effects? Are there any references for this? In such a case, the Henry's law constants for pure water may not be applicable.

"After addition, the lowest measured initial liquid phase concentration was 17.8 mg/L (11% decrease)." Is this an expected trend or is this random variation due to evaporation of acids?

Response:

Thank you for pointing it out. Perhaps these sentences might create more confusion than clarity to the readers. We also did not refer to the salting effects either. We meant to say that the addition of HCl and NaOH didn't create a bias in terms of concentration in our measurements. We measured the solute concentration before freezing, after freezing and desorption, for calculating retention coefficients. So, the changes in mass concentrations are accounted for already. As such, the referred texts in L219-224 have been removed altogether to avoid any further confusion. The changes can be seen in the track-changes document.

Editor:Table 3: Please indicate that you use dimensionless Henry's law constants.

Response:

Text has been changed in Table 3 as:

"Dimensionless Henry's law constant" in place of previously written "Effective Henry's law constant"

Editor:l. 325/6: '*from their numerical simulations'* seems at a wrong place in the sentence. Please clarify.

Response:

Text has been changed for better clarity in L326 as:

"Stuart and Jacobson (2006) reported the formation of liquid pockets that can trap solutes during freezing, informed from previous studies of dendritic crystal growth in solutions."

Editor:l. 343: "*Our results show higher retention coefficients close to 1 for mm sized raindrops for similar substances from previously studied retention coefficients"*

This sentence should be restructured for clarity, e.g. Our results show higher retention coefficients (close to 1) for similar substances in mm sized raindrops as compared to previously determined retention coefficients in um sized cloud droplets.

Response:

Thank you for your suggestion. Text has been changed in L365 as:

"Our results show higher retention coefficients (close to 1) for similar substances in mm sized raindrops as compared to previously determined retention coefficients in μm sized cloud droplets (von Blohn et al. 2011; Jost et al. 2017; Borchers et al. 2024)"

Editor:Section 4: Please make sure that the conclusion section adheres to the author guidelines at https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html

Response:

Thank you for pointing it out. We have added a summary section, which was missing in our conclusions. And rearranged the conclusions as per the guidelines provided.

Text has been added in L350 as:

"At the onset, we successfully characterized the freezing of levitated rain drops (2.0 ± 0.1mm) at three different concentrations and temperatures using the acoustic levitator setup. We measured the retention coefficients of nitric acid, formic acid, acetic acid and 2-nitrophenol as single components and their combinations as binary mixtures, during the freezing of rain drops. In addition to these measurements, we also checked the sensitivity at three different pH levels (pH 3, 4 and 6/7) and at two different temperatures (−3.9 ± 0.3◦C and −6.9 ± 1.1◦C)."

References:

Borchers, C., Seymore, J., Gautam, M., Dörholt, K., Müller, Y., Arndt, A., Gömmer, L., Ungeheuer, F., Szakáll, M., Borrmann, S., et al.:Retention of α-pinene oxidation products and nitro-aromatic compounds during riming, Atmospheric Chemistry and Physics, 24, 13 961–13 974, 2024.

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