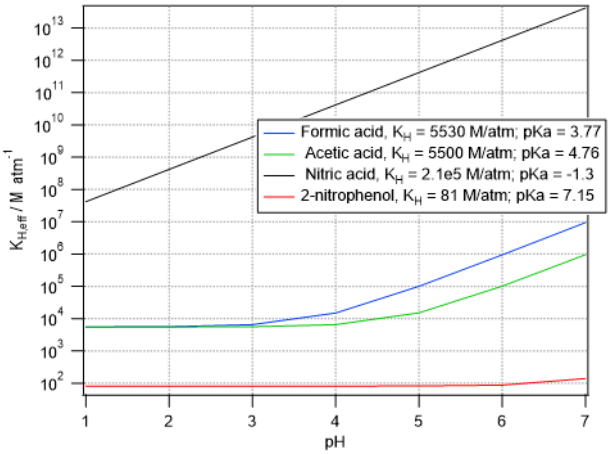
Main editor comments: (Line numbers refer to the manuscript version with track-change).

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.

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 (K_H*) 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.



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.

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.

Minor/Technical comments

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

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 …

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

l. 61: Freezing of raindrops is…

Table 2: …temperature was

l. 323: …rates … imply

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…

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'?

l. 27: 'evident' seems redundant here

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

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

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.

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’.

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?

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

l. 69: what do you mean by ‘…values for riming with cloud droplet sizes’ – is it simply ‘in riming cloud droplets’?

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’

l. 111: ‘least’ should be ‘lowest’

l. 120: ‘benzoic’ misspelled

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

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’.

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

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

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

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.

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

l. 193” replace ‘least’ by ‘lowest’

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?

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?

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

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

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.

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