Authors' responses to the Co-Editor comments

First, we would like to thank the Co-Editor for her thorough revision and valuable comments.

In the following, the Co-Editor's comments or questions are written in bold font, our answers in standard font, and the changes within the manuscript in italic font.

Technical Comments

1. All grammatical errors have been corrected according to the Co-Editors suggestions.
2. The notation OH_X was an oversight and has been changed to HO_X on page 2 in lines 25 and 29.

Specific Comments

1. Page 8, line 12: Why not -12°C?

We chose the temperature of -11.5°C, because it corresponds to the mean temperature of all measurements.

2. Page 10, line 16: Isn't this energy already accounted for in the thermodynamic constants used to derive H*? An explanation that relies on kinetics seems more plausible to me.

According to the Co-Editors comment the argumentation has been modified.

Even though association (recombination) occurs quickly compared to the other timescales involved in the retention process (e.g., those of aqueous phase transport, interfacial transport, gas phase transport of a molecule, and the freezing time), it influences the retention of acetic acid less than that of formic acid. This is because acetic acid is three times more present in the molecular form compared to formic acid, which facilitates its escape to the gas phase. Furthermore, the association timescale for acetic acid is one order of magnitude faster than formic acid, which further increase the degassing rate for acetic acid or, on the other hand, decrease that for formic acid.

3. Page 11, line 15: The correct unit should be used.

Unfortunately the formulation was a bit misleading. Here the fraction of the total formaldehyde which is present as methanediol or as monomeric formaldehyde was meant. For the sake of clarity we rephrased the sentence as follows:

Especially at low concentrations, the –diol form is the favored one (Walker, 1964). According to the hydration constant, K_{hyd}, at T= 298 K 99.9% of the total dissolved formaldehyde is present as methanediol, whereas less than 0.1 % is present as monomeric formaldehyde.

4. Page 11, line 25: If your effective Henry's Law constant H* already takes into account the fast hydration and slow dehydration, through the equilibrium constant for R1, then this is not a separate explanation.

Our argument is as follows: The solution for the droplets is initially in equilibrium according to the effective Henry's law constant (including K_{hyd}). After the injection into the wind tunnel and during freezing the droplets experience non-equilibrium conditions which are not determined by the effective Henry's law constant anymore. The non-equilibrium conditions are induced by a low ambient gas concentration, the temperature increase of the supercooled droplets to 0°C during freezing, and the build-up of a radial concentration gradient inside the droplet induced by the segregation of the molecules from the developing ice. All these processes shift the equilibrium towards the gas phase but with a rate constant determined by k_{-R1}.

5. Page 11, line 27: Since you measure the aqueous phase formaldehyde by converting to formic acid, is it possible that some of the formaldehyde is actually oxidized within the super-cooled water or ice (before your addition of H2O2) and that explains the apparently high retention coefficient?

In our opinion this was unlikely to occur, since the total formaldehyde concentration in aqueous solution is several orders of magnitude higher than, for example, the OH concentration. Even absorbed and dissolved H_2O_2 would not have been able to oxidize parts of formaldehyde to formic acid since the reaction rate is really low in the pH range of the droplets. Especially because the exposure time in the wind tunnel air was just about 8 s. Moreover, the oxidation to formic acid would actually have decreased the retention coefficient of formaldehyde, since the retention coefficient of formic acid is lower than 1. Consequently, if some part would have been oxidized to formic acid prior to riming this would cause a retention coefficient of formaldehyde lower than 1.