Reviewer #3

We thank Reviewer #3 for his or her useful comments for this manuscript. We address specific points below.

MAJOR CONCERNS

The use of X (chi) to represent good fit and mass fraction in the text is somewhat ambiguous. Perhaps the author could use different variables.

We chose and 2 for these two parameters based on the original work in each case, but we agree with the reviewer that this leads to some ambiguity. We now use 2 to indicate the goodness of fit and to describe the weighting term of the Szyszkowski-Langmuir equation for clarity.

Each droplet is allowed to equilibrate over 2-5 minutes. On average, how long did it take to perform a set of experiments on a given sample? Both Formaldehyde and acetaldehyde have low vapor pressures at room temperature. Would the evaporation of these compounds during measurement effect surface tension measurements? Has this been considered in the measuring protocol? It is mentioned in Table 1 that for formaldehyde mixtures the organic concentration was variable but little to no discussion is given in the text.

Experiments on a given sample typically lasted ~ 1 hour. In a previous study, we tested both formaldehyde and acetaldehyde in aqueous and ionic solutions (see Li et al., 2011) via the same methodology and equivalent sampling times, and showed that while both species are volatile, the surface tension decreases with increasing organic concentration in the bulk aqueous solutions. This indicates that enough of the organic remains present in the aqueous phase (either by becoming less volatile when hydrated or forming oligomers) to suppress surface tension over the time periods that we used for this study. We have not taken volatility effects of these organics into account in terms of the supplied organic concentrations.

The variable organic concentrations of formaldehyde mixtures in Table 1 indicate variable aqueous organic concentrations; we have updated Table 1 and its caption to be clearer.

MINOR CONCERNS.

Figures. This reviewer suggests restating the relevance of the black and gray lines in at least one of the figure captions.

We have restated this information within the first figure caption of the manuscript and the supplement.

P555 L1. What is the conductivity of the Millipore water used?

We use a resistivity of 18.2 M \cdot cm for Millipore water, equal to a conductivity of 0.055 μ S/cm.