

## Referee #1

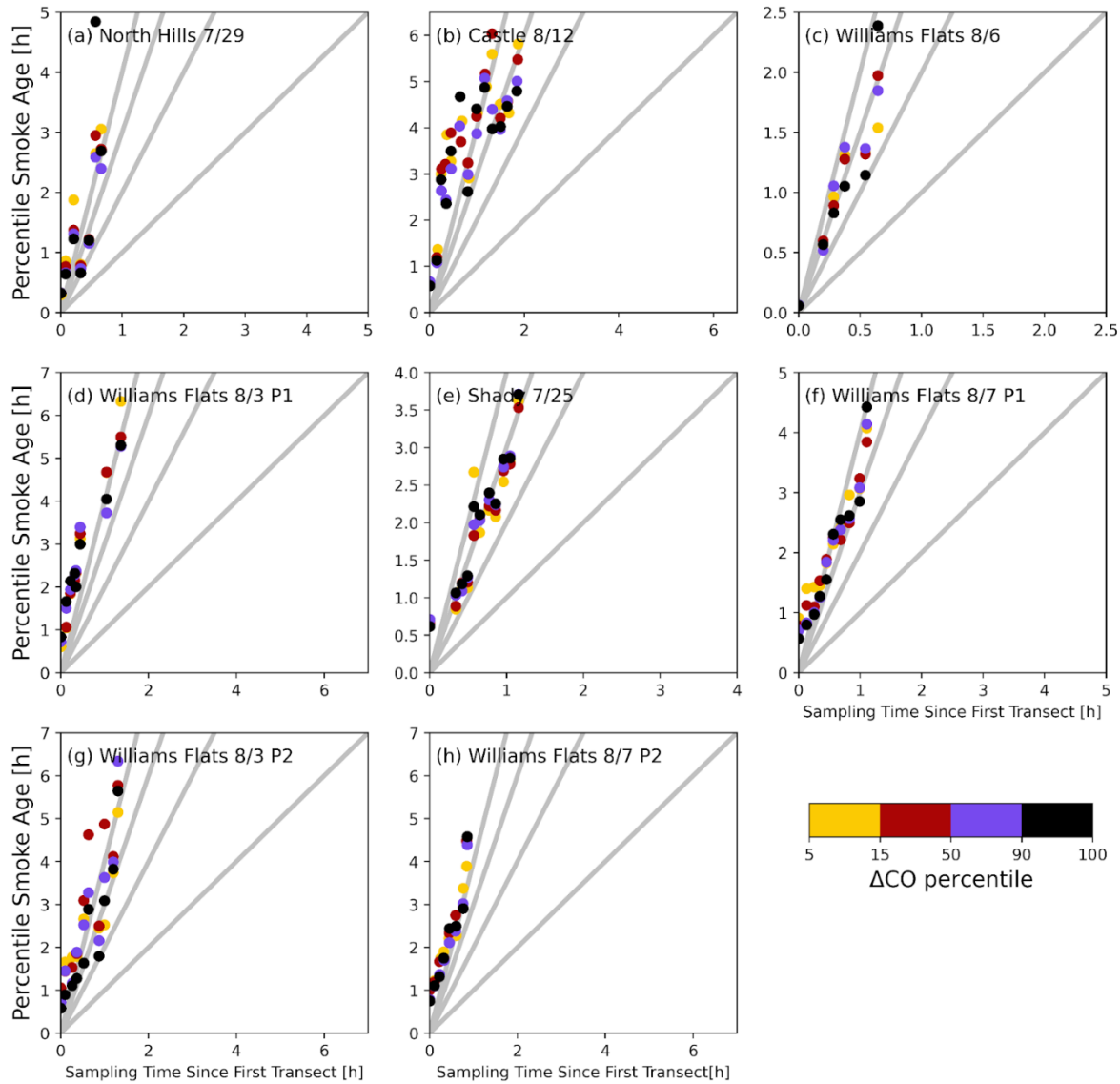
This work used aircraft measurement and box model simulation investigated the impact of plume concentration on coagulation and OA condensation/evaporation and the associated impact on particle growth during FIREX-AQ. The authors emphasized that one of the highlights of this work is using a large number of pseudo-Lagrangian samples to investigate this topic. However, uncertainties of pseudo-Lagrangian samples were not well investigated. Unexpected performance of field data and disagreement between observation and simulation were attributed to pseudo-Lagrangian samples which weaken the conclusions made by the authors.

For major question 1, I am still not be convinced by the explanation on smoke age. Although previous studies used the straight line horizontal advection definition of smoke age in their works, they did not investigate in-plume gradients in much greater detail as that done by the authors in this work. I would like to see more discussions on why ‘these uncertainties are likely smaller than the uncertainty due to issues of imperfect Lagrangian aircraft sampling’.

*We have modified the sentence quoted just above by the reviewer to reflect less confidence in the relative uncertainties: “In addition to the uncertainties from the pseudo-Lagrangian sampling, there are likely uncertainties in the smoke age due to the wind shifting directions and potentially wind velocity varying radially within the plume (discussed more later in this section).”*

*Additionally, we have tested how much the calculation of age may vary between the percentile bins. In the methods paragraph detailing the splitting of the transect into percentile bins to examine in-plume gradients we have added discussion on these calculations as well as an SI figure (Figure S7).*

*“The mixing may also mean that there are differences in the smoke age in the percentile bins due to the time for the initial momentum of the smoke plume to equilibrate with the velocity of the environmental air at the injection level. In Figure S7, we show the ages of each percentile bin for each transect derived separately using the mean wind speeds in the percentile bins and the distance from the fire. While the derived ages vary by around 20 to 30 minutes between the 5 to 15 and 90 to 100 percentile bins, there are no systematic differences with one bin being generally younger or older than the other. Further, the difference in the plane speed and wind speeds cause the imperfection in Lagrangian sampling to be larger than the variability in the smoke age in the percentiles. Therefore, we use the single value of smoke age for each transect included in the dataset for both the transect average and percentile bins.”*



**Figure S7:** The physical smoke age in the percentiles versus the sampling time since the first transect in seconds for each of the eight sets of transects. The gray lines have slopes of 4, 3 and 1, with the 1:1 line representing the ideal slope for Lagrangian sampling.

For major question 2, the authors misread my question. I did not say OA changes are predicted by the model. I meant the size changes due to coagulation, dilution and adjust for OA condensation/evaporation were simulated by the box model. The authors found the changes of the simulated diameter growth by OA condensation/evaporation (Line499-501) were small, so they concluded that OA evaporation/condensation having a relatively minor impact on the diameter growth. It needs to be noted that this conclusion is based on box model simulation done in this work. Therefore, it is worth checking the representation of aerosol microphysics processes by this box model based on information presented in the paper. As shown in Fig.7a and b, simulated diameters with adjustment for OA condensation/evaporation are larger than those without the adjustment. It means the adjustment is dominated by OA condensation. By contrast, as shown in Fig.7c, d, f and g, simulated diameters with adjustment for OA condensation/evaporation are smaller than those without the adjustment. It means the adjustment is dominated by OA evaporation. These are agreed with the information presented

in Fig.S8. Things puzzled me is that I thought the domination of OA condensation/evaporation is changing with smoke age as shown by dots in Fig.S8. But I can only see continues OA condensation in Fig.7a and b and continues OA evaporation in Fig.7c, d, f and g. The authors need to clarify why model simulation does not reflect the variance of OA condensation/evaporation with smoke age shown in Fig.S8.

*We believe that the reviewer is confused by the method we use for specifying the  $D_p$  change due to OAER changes in the model. We state in the manuscript, "OA condensation/evaporation was included in the model based on the observed **trends** in OA (Fig. 5c) and Eq. 6.", rather than including the exact OAER for every transect. To make this point more clear we have added the following sentence, "We are basing OA condensation/evaporation on the linear fit of the points in Fig. S11, and there does not appear to be any systematic change in the slope as any of the plumes age." Note that Fig. S11 in that quotation is the Fig. S8 referred to by the referee. We edit the next sentence to reference the main text figure for OAER trends "Consistent with the OAER trends (Fig. 5c), net condensation grows the particles in comparison to the coagulation-only model diameter in 3 cases, and net evaporation shrinks the model particle diameter in 5 cases (Fig. 7)." Additionally, we edit the methods section following Eq. 6 to say "Where  $d(OA/CO)/dt$  is the average observed change in the OA enhancement ratio with time from an ordinary least squares regression, and  $t$  is the simulation time." This clarification in the methods section is to emphasize we are using the average rate of change of OAER to constrain condensation/evaporation, not the difference in OAER from one transect to the next.*