Referee #2

We thank the referee for their time and helpful comments.

This manuscript presents airborne observations of some aerosol parameters (aerosol particle median diameter, number concentration of particles > 100 nm, organic aerosol enhancement ratio (OAER, deltaOA/deltaCO) and O:C ratio) as function of atmospheric age for eight major wildfire plumes in the western US in 2019. The main finding of the manuscript is that the median diameter of particles > 100 nm increases faster in plumes that have higher OA concentration. A box model suggests that this change is mainly due to coagulation, OA evaporation and condensation having a minor role.

Biomass burning aerosol size distribution and its evolution in sub-grid scale is an important topic for climate and air quality. However, it is unclear what are the novel findings in this study and there are also some major comments need to be addressed before this manuscript can be accepted.

There are several novel aspects of our study, and we have now added text to point these out more explicitly. While previous studies have concluded that coagulation explains the majority aerosol number size distribution changes, we're unaware of a study that has investigated this for a large number of pseudo-Lagrangian samples. We show in field data that coagulation dominates the aerosol size distribution changes, this had only been speculated in prior field data and shown in theoretical studies. We also investigate in-plume gradients in much greater detail than the Hodshire et al. (2021) study, showing relationships between plume concentration and plume evolution in both transect averaged and in gradients within the plume. We show through modeling and calculated mixing times that the mixing within the plume is generally too fast to be able to apply the relationships derived from in-plume gradients to plumes of a similar average concentration. Mixing was not considered in the prior Hodshire study investigating in-plume gradients in this way. The limitations of prior studies are discussed in the introduction. We have added to the final paragraph of the introduction lines stating what is new when we overview the work of this manuscript.

"We use an aerosol-microphysics model to estimate how much of the aerosol size growth is due to coagulation versus OA condensation/evaporation; the first study to show in multiple Pseudo-lagrangian transects of smoke plumes the dominance of coagulation. Finally, we investigate the timescale of mixing between the more and less concentrated regions of plumes to determine if aging in these portions of the plumes can be assumed to occur independently; prior studies have not investigated this role of mixing."

Major comments

The evolution of the size distribution is described through two parameters only: median diameter and number concentration. Furthermore, the size distribution is available only for particles > 100 nm in diameter - were there really no measurements below 100 nm? Even for particles > 100 nm, please show the actual size distributions. Does the initial size distribution depend on OA concentration? Does the width of the size distribution agree with the modelled time evolution? Is one mode enough to describe the size distribution or should more modes be used?

FIREX-AQ did have a SMPS on the aircraft to measure particles smaller than 100 nm. However, the SMPS scans take 60 s, during which the aircraft traveled approximately 8 km, this makes it too slow to use in our analysis. Moore et al. (2021) showed that for points when the SMPS was scanning the plume, the LAS captured the majority of the mode, and there was not a clear, smaller Aitken mode.

We have added a SI Figure (Fig. S5) to show the measured size distributions along with the lognormal fits of the size distribution for every transect for each of the eight cases. We believe that based on the shown fits of the LAS binned size distribution, that one mode is enough to describe the size distribution (as well as the findings in Moore et al. (2021)). Additionally, the use of the fit allows us to estimate particles in the tail of the distribution below 100 nm.

Investigating modal width is an excellent suggestion. We have changed Fig. 3 to now be a 6 panel plot showing the initial diameter, width, and number enhancement ratio as a function of OA concentration for the transect averages and ΔCO percentiles. All of these variables are related to the initial OA concentration, we have added a paragraph discussing this. And Fig. 4 is now the average rate of change of median diameter, modal width, and number enhancement ratio as a function of OA concentration in both the transect averages and ΔCO percentiles. We have added discussion on the rate of change of modal width.

We have added an SI Figure (Fig. S16) that showing the comparison between modeled and observed width. The model captures some, but not all of the reduction in the width. As well, Fig. S17 has been added to show the normalized initial and final size distributions in the model and observations.

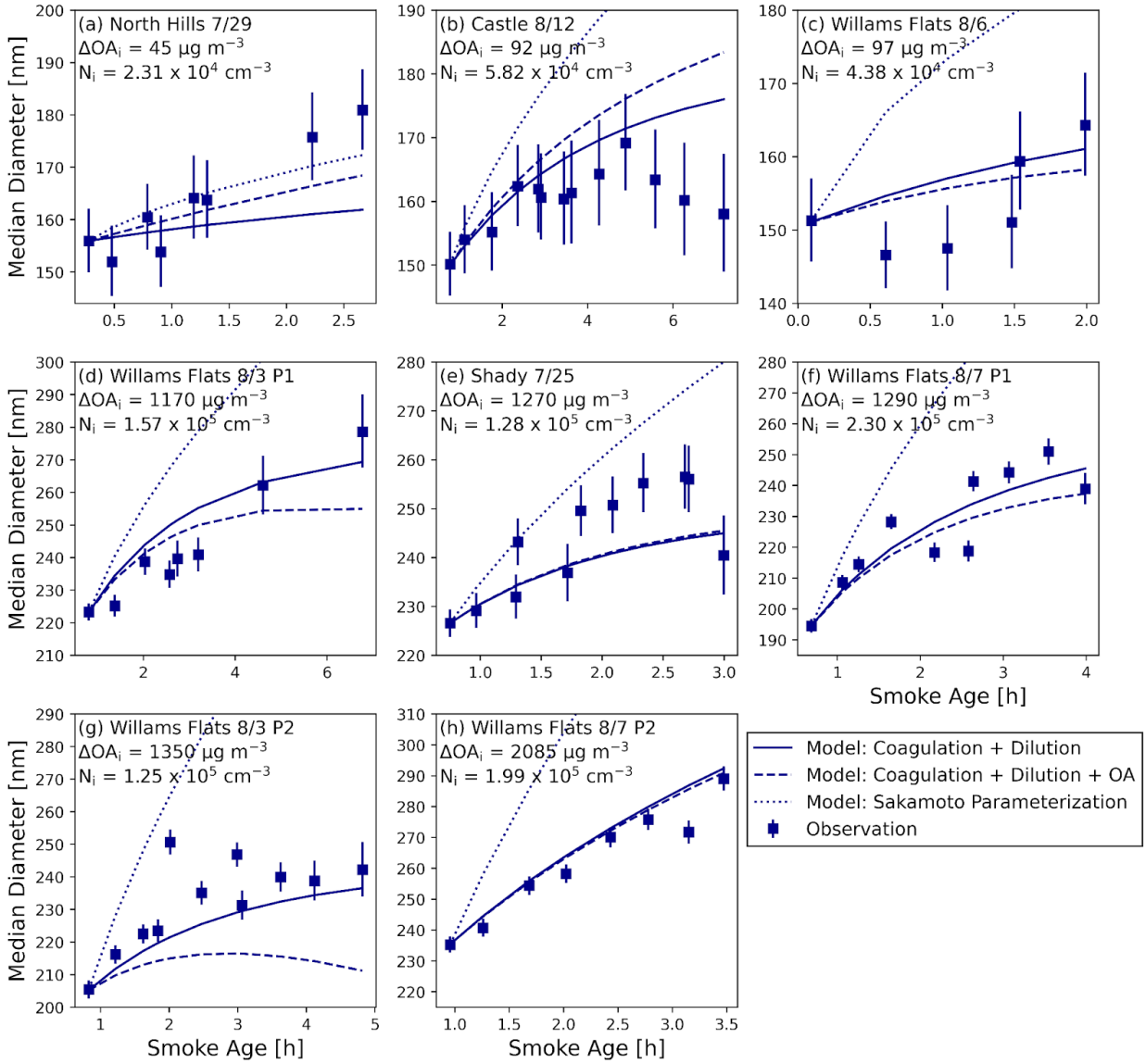
It seems that most deviations from the modelled time evolution (e.g. Fig. 7) are explained by sampling not being Lagrangian. Please try to identify the processes that cause these deviations and discuss them in more detail. Can the model be used to evaluate whether changes in dilution and photochemical age would be enough to explain the observed differences, or if the differences are due to other factors (e.g. emission changes)?

In our discussion, we have added, "Non-Lagrangian sampling may be impacting the observations through a plume injection height change, so the aircraft is no longer sampling the same vertical location of the plume; or an emissions factor or fire radiative power change due to the diurnal cycle of fires."

In its current state, the model cannot be used to evaluate whether time changes in dilution, age, and emissions would be enough to explain the observed differences. It would also be difficult to do this without a true Lagrangian set of transects to compare to. In our results section, we suggest ideas for future field studies to help to understand the impact of non-Lagrangian sampling.

The coagulation parameterisation by Sakamoto et al. (2016) is mentioned (e.g. lines 85-92), but no comparison is made. Please evaluate the Sakamoto et al. (2016) parameterisation with your observations.

*The parameterization is difficult to initialize for the flights since it is based on a mass flux*fire area/wind speed/mixing depth (dM/dxdz), which includes emission rates that we do not have. We estimate the dM/dxdz value by integrating the particulate matter concentrations across each transect; however, due to the aircraft attempting to sample the plume generally near its most concentrated point vertically, the estimated value is likely biased high relative to the average value in the plume. As shown in the figure below, this results in the Sakamoto parameterization being biased high. However, the Sakamoto parameterization does show more growth in the concentrated plumes, so qualitatively it follows expectations. We do not add the Sakamoto results to the main text or SI, since we are unable to represent dM/dxdz in the way that was intended by the parameterization.*



Minor comments

On line 254 “N” is used for nitrogen, which could be confusing as it is used for number concentration lines 251 and 257.

Wrote out Nitrogen instead of using N.

line 258 “N is the number concentration between 50 nm and 800 nm, the range of diameters used to fit the dN/dlogDp measurements.” So are you showing only the fitted N, and not the directly measured parameter? Please use the measured quantity in the figures.

As noted in the text, we use the fitted quantity to capture the tails of the size distribution. We have continued to use this fitted value in initializing the model, and in Fig. 4, Fig. S9 (Formerly S7), and Fig. S15 (Formerly S13). The measured quantity of each bin can be seen in the new supplemental figure Fig. S5, showing the full size distributions with age for each case. We also have expanded the range of the fit to be between 50 nm and 2000 nm.

line 262 “number enhancement ratio” defined for the 2nd time.

We have removed this second definition.

line 293 I believe the Pasquill stability classes are defined for boundary layer, including convective mixing. Can this method be applied to mixing above boundary layer, where the measurements were done (line 174-175)?

We are extrapolating to above the boundary layer in this procedure, and we now explicitly state this in manuscript, “As a check on the mixing time calculated from the stability class, since we are extrapolating the Pasquill stability class to above the planetary boundary layer, we also calculate a mixing time from the rate of change of the ΔCO gradient between the core and edge regions.” Also we estimated the stability class two ways, but the second way was only discussed briefly in the main text with results in this SI. The alternate way to estimate the mixing time uses the rate of change of the CO gradient between the core and edge regions of the plume. These two methods yielded similar results, and both showed that the majority of the plumes mixed in <5 hours with a single plume (Williams Flats 8/7 P2) taking >10 hours to mix. This general agreement provides confidence in our conclusion that most of the plumes are mixing on timescales similar to the sampled aging time with the exception of Williams Flats 8/7 P2. We have added a column to Table 2 to include the mixing times derived from the CO-gradient method, as well as more discussion related to the CO-gradient method.

line 322-323 “Additionally, this assumes volume-controlled growth/shrinkage, where all particle sizes grow/shrink by the same fractional amount, preserving the lognormal modal width.” Examining the initial size distribution as function of OA concentration could help validate this assumption. At least for the median diameter (Fig. 2), there seems to be a clear distinction between more diluted plumes (Dpm ~150nm) and the more concentrated plumes (Dpm ~200nm).

Thanks for this suggestion. We now include initial size distribution properties as Fig. 3 that show a clear relationship between the initial diameter and initial OA. In our discussion of the initial OAER values in Fig. 5, we now discuss that the smaller diameters in diluted plumes can partially be explained by the lower OAER in the dilute plumes (with coagulation prior to the first transect likely explaining the rest), with both indicating faster evaporation prior to the first measurement in the dilute plumes.

"The lower initial D_{pm} in dilute plumes (Fig. 3a) also suggests faster evaporation. Between a ΔOA_i of 100 µg m^{-3} and 1000 µg m^{-3}, initial OAER decreases by a factor of about 0.62, which if only evaporation was occuring would suggest the particles in the 100 µg m^{-3} plume would be 0.85 times smaller assuming the emitted diameter is not correlated with concentration at the first transect. We observe particles that are 0.76 times smaller at a ΔOA_i of 100 µg m^{-3} compared to 1000 µg m^{-3} (Fig. 3a), suggesting that evaporation prior to the first transect is contributing to smaller particle sizes for less-concentrated plumes."

line 341-343 "Castle 8/12 also has a larger uncertainty range due to a constant increase in Dpm for the first five hours of aging, but then a decrease in Dpm during the final three transects, potentially due to deviation from Lagrangian sampling." Can you use the size distributions to rule out growth of < 100 nm particles into the sizes where they can observed?

Based on the size distributions shown in Fig. S5, there does not appear to be an increase in the number of particles in the smallest size bins. Therefore, it is unlikely that particles smaller than 100 nm are growing into the sizes where they can be observed.

line 428 Do you mean Fig. 6a-b?

No, this reference is meant to be referring to the initial OAER figure, which is Fig. 5a-b in the original manuscript on ACPD.

Figure S2 Please check x-axis label.

Fixed x-axis label to be Smoke Age [h].