This study reports characteristics of fast time-dependent particle size distribution from lab scale biomass fires for southwestern US fuels. The study was carried out in the combustion facility at the USDA Forest Service's Fire Science Laboratory (FSL), Missoula, MT. The study claims to be the first of its kind in providing details regarding the evolution of particle size distribution in wildland fires, and a way to separate out size distribution characteristics with respect to the flame Modified Combustion Efficiency (MCE).

Major Concerns:

1) The study uses two different sizing techniques to measure particle size – mobility and aerodynamic. However, no where in the manuscript do the authors define and/or briefly introduce – a) the basic concepts and equations governing the two sizing techniques, and b) the working of the FMPS and the APS. I strongly suggest the authors to also include citations when introducing these concepts in the manuscript. OP-FTIR spectrometer also requires a brief introduction and citations for readers' convenience.

- ⇨ Below will be added in the revised draft.
- ⇨ The Fast Mobility Particle Sizer Spectrometer (FMPS, TSI Model 3091) is capable of measuring mobility diameter of the particles. The instrument is made of two concentric cylinders (Classification columns), a corona diffusion charger, and 32 electrometers, which covers range of 5nm to 560nm. The stream containing positively charged particles flows along with sheath air. The high voltage between the two cylinders carries the particles from the side that they are introduced to the other side that has the electrometers. Close the top of the column, the particles with higher electrical mobility are collected, and further upstream the particles with lower electrical mobility are collected (TSI manual). Electrical mobility, Z is defined as below

$$Z = \frac{neC}{3\pi\mu_g D_m}$$

Where n, e, C, μ_g, D_m are number of charge, charge of an electron, slip correction factor, gas viscosity and particle mobility diameter.

The aerodynamic diameter D_a is the diameter of the unit density ($\rho_p = 1\text{g/cm}^3$) sphere that has the same settling velocity as the particle (Hinds, 1982). The Aerodynamic Particle Sizer (APS, TSI Model 3321) is capable of measuring particle size distributions in the size range of 0.5 and 20 µm. The sample particles are accelerated as the carrier gas flows through a converging nozzle. Due to the inertia, particles cannot accelerate as fast as the gas and the velocity lags that of the gas. Particle size is related to this velocity lag and that is measured at the nozzle exit when particles pass through two partially overlapping laser beams. The time of flight between the two laser beams is then used to calculate the aerodynamic diameter (Rader et al, 1990; Wang and John, 1987). Chen et al. (1985) found a unique calibration curve exist between particle Stokes number and normalized particle velocity against the gas velocity.

The Stokes number is defined as

$$St = \frac{\rho_p D_p^2 V_g C}{9\mu_g W}$$

where ρ_p, D_p, V_g, C, μ_g, W are particle density, particle diameter, gas velocity, slip correction factor, gas viscosity and diameter of the nozzle at the exit. The APS measured aerodynamic diameter requires correction for non-Stokesian effects when particle Reynolds diameter becomes greater than 0.5 (Wang and John, 1987). A conversion of aerodynamic diameter into mobility diameter requires a correction factor involving dynamic shape factor and density of particles (Stöber, 1971). Current study does not aim to measure those parameters for conversion.

⇨ Every molecule has its own specific IR radiation absorption pattern. Researchers have used this fact in Fast Fourier InfraRed(FTIR) spectroscopy. If a beam of IR is passed through smoke, and the IR spectrum is recorded, the acquired graph will have peaks that are due to molecules present in the smoke (Yokelson et al. 1997, Goode et al. 1999). The OP-FTIR instrument used in this study included a Bruker Matrix-M IR Cube spectrometer and a thermally stable open white cell. The white cell was positioned on the sampling platform approximately 17 m above the fuel bed so that the open white cell spanned the stack directly in the rising emissions stream. The white cell path length was set to 58 m. Several extensive tests were performed to optimize the many sampling parameters, including duty cycle, sample frequency, and spectrometer options and chose a spectral resolution of 0.67 cm^{-1} and spectra were acquired every 1.5 seconds (four co-added spectra in 1.5 seconds) beginning several minutes prior to the fire and continuously until the end of the fire. A pressure transducer and two temperature sensors were located adjacent to the optical path and were logged on the instrument computer and used for spectral analysis.

2) The study fails to convince the reviewer about the reliability of the measured size distributions. It is a well accepted fact that flaming combustion produces fractal-like particles. Fractal-like particles will show significant differences in the size distributions measured by mobility and aerodynamic techniques. These are not real discrepancies; instead, they merely capture the different dependence of both equivalent diameters on the fundamental particle properties. This phenomenon has been observed in the measurement of ambient aerosol. Please refer to (Chakrabarti, B., Singh, M., and Sioutas, C. (2004). Development of a Near Continuous Monitor for Measurment of the Sub-150 nm PM Mass Concentration,Aerosol Sci. Technol. 38(S1):239–252.) That said, I would like the authors to provide some basic information on:

⇨ The size range of fractal-like particle generated by combustion process is usually below 400nm in mobility diameter. Fractal-like particles bigger than 400nm constitute so-called coarse mode. These particles are formed by particles re-entrained from exhaust walls. We have not observed significant number of fractal-like particles in the size range that the APS (Aerodynamic particle sizer) measures. Particles in the range the APS measured were mostly tar ball or ash particles from our supplemental TEM measurement which is not reported in this draft.

⇨ As the measurement range of the two instruments (FMPS and APS) has minimal overlaps, we do not aim to convert aerodynamic diameter to mobility diameter. Acquiring shape factor requires mobility and aerodynamic measurement to be done in series not in parallel.

a) Whether any charge correction was used when measuring the mobility size distribution of the particles. It is very likely that multiple charges could affect the size distribution retrievals of the already charged smoke particles. This could cause error in determining the peak of the particle geometric mean diameter.

- ⇨ Yes, the charge correction was performed for the data we have presented in the original draft.

b) A logical explanation as to why the major modes of their size distribution as measured by the FMPS are smaller than what has been observed by previous studies conducted in the USDA FSL (e.g. Chakrabarty et al. (2006)).

- ⇨ Chakrabarty et al. (2006) reported particle size distribution for wet and dry fuel conditions. Major mode diameter for the dry fuel ranges 40-45nm for three burns out of six burns (see their figure 12). Therefore their results are similar to ours. Our fuels were dry in most of cases.

What was the flow regime of these particles? Flow regime is very important information as it helps determine the dynamic shape factor information of the particle. The dynamic shape factor directly affects the particle mobility diameters. For e.g. a fractal-like particle experiences a larger drag force but the same electrical force compared to its volume equivalent sphere, so it is “sized” as a mobility-equivalent sphere that is larger than its volume-equivalent sphere. I strongly suggest the authors to think on these lines and justify their observation. This will give clarity to the readers.

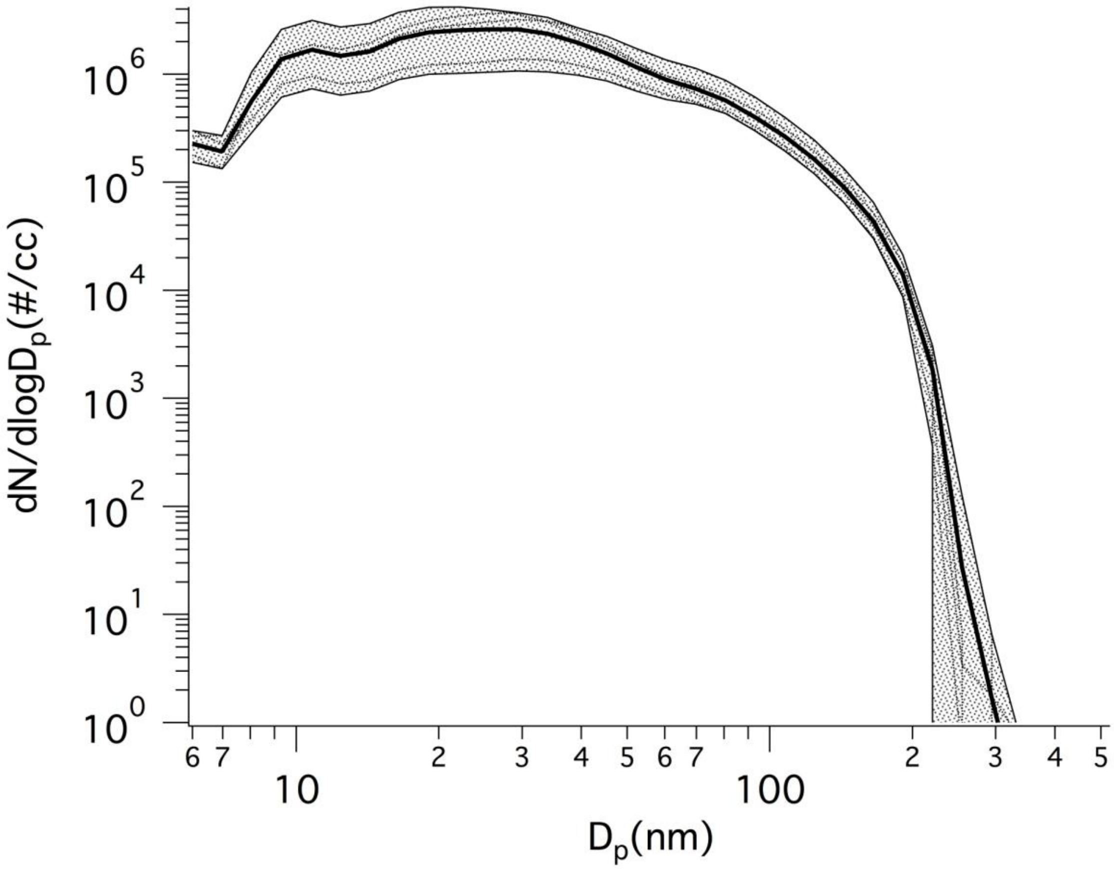
- ⇨ The particle Reynolds numbers are reported as 0.65, 4.8, 24 and 103 for diameters of 1.0, 2.1, 5, and 15um by Wang and John (1987) for the APS at the nozzle flow velocity of 150 m/s. We have also confirmed with TSI (personal communication) that Wang and John (1987)’s Reynolds number is valid for the latest APS model (3321) which we have used for this study.
- ⇨ Since the fraction of fractal-like particles in the size range the APS measured is insignificant for the reasons mentioned above, the correction for the fractal like particles was not considered.

3) The distinguishing of the modes of combustion using slopes of MCE vs. geometric mean diameter fails to provide any insight to the readers. Why should this technique prevail over the commonly used technique of using only MCE values?

- ⇨ Ward and Radke’s MCE criterion gives a guideline to distinguish different phase of combustion but it fails to give accurate distinction at specific MCE value. For example smoldering mode starts at different MCE value of 0.82, 0.76 and 0.83 for different burns at Fig 9abc. As the combustion of biomass is always dynamic instead of being steady state, either combustion or emissions parameters should evolve at different rate and direction at different phase of combustion. When we plotted MCE value against geometric mean diameter, we found that the distinction between different phases of combustions could be more clearly seen. We converted the original plot to the color plot to present time evolution of MCE and geometric diameter better.

First off, there has been no error analysis done for the size distribution measurements done in this manuscript.

- ⇨ Figure 4 and 5 are burn averaged size distributions. Below figure shows error bounds for FMPS number distribution for the fuel of oak savanna. Thick solid line represents average of six burns and shaded area shows variation.



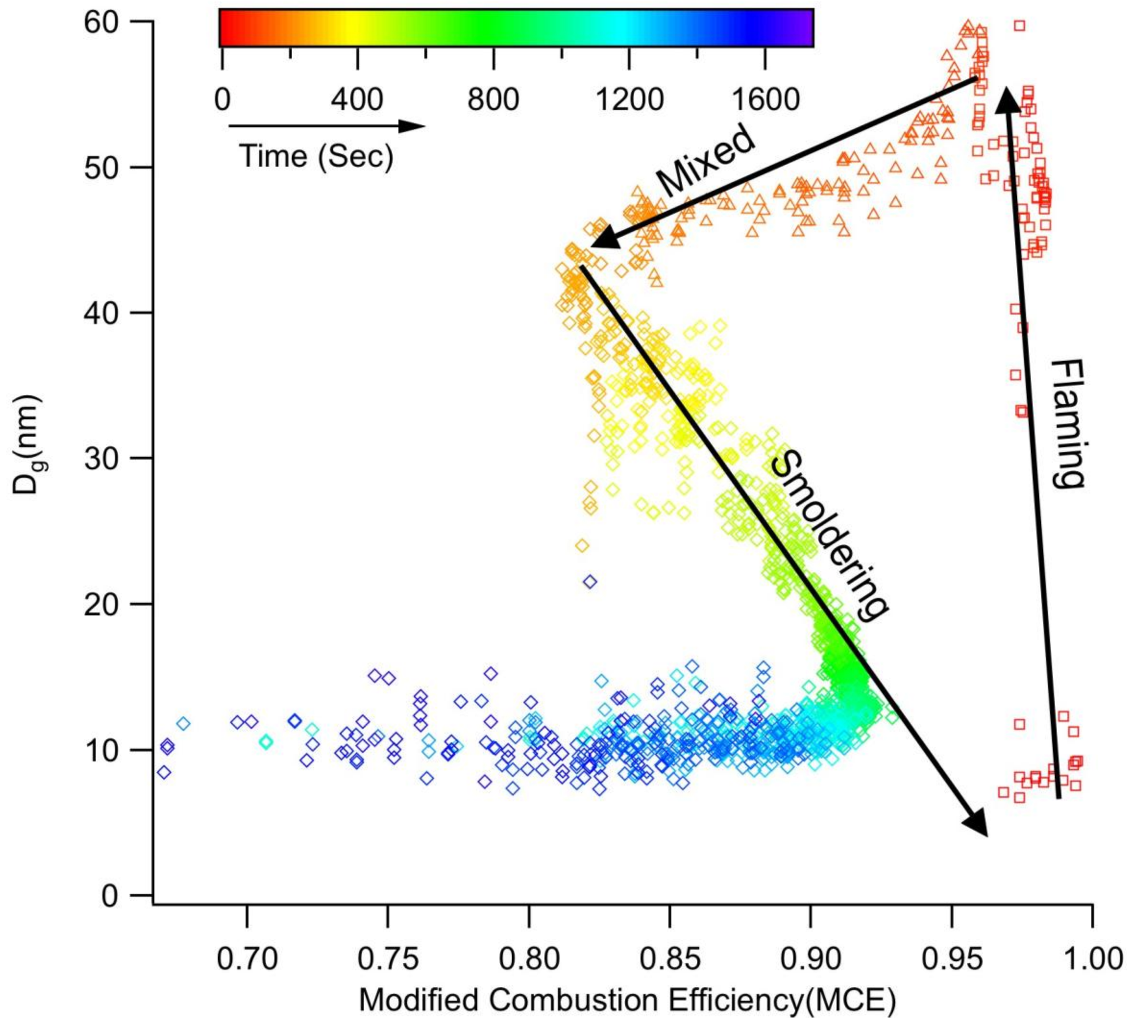
⇨

Secondly, the authors never provide any convincing argument – laid on
a strong theoretical foundation – regarding the validity and reliability of their measurements.
In Fig. 9(c)., the trend of the graph is different than that of Fig. 9(a) and (b).
Why?

- ⇨ That is because the MCE and geometric diameter vary at different rate at different burn. Also this is why we claim that our MCE vs geometric mean diameter can be a useful graph to distinguish different phases of combustion.

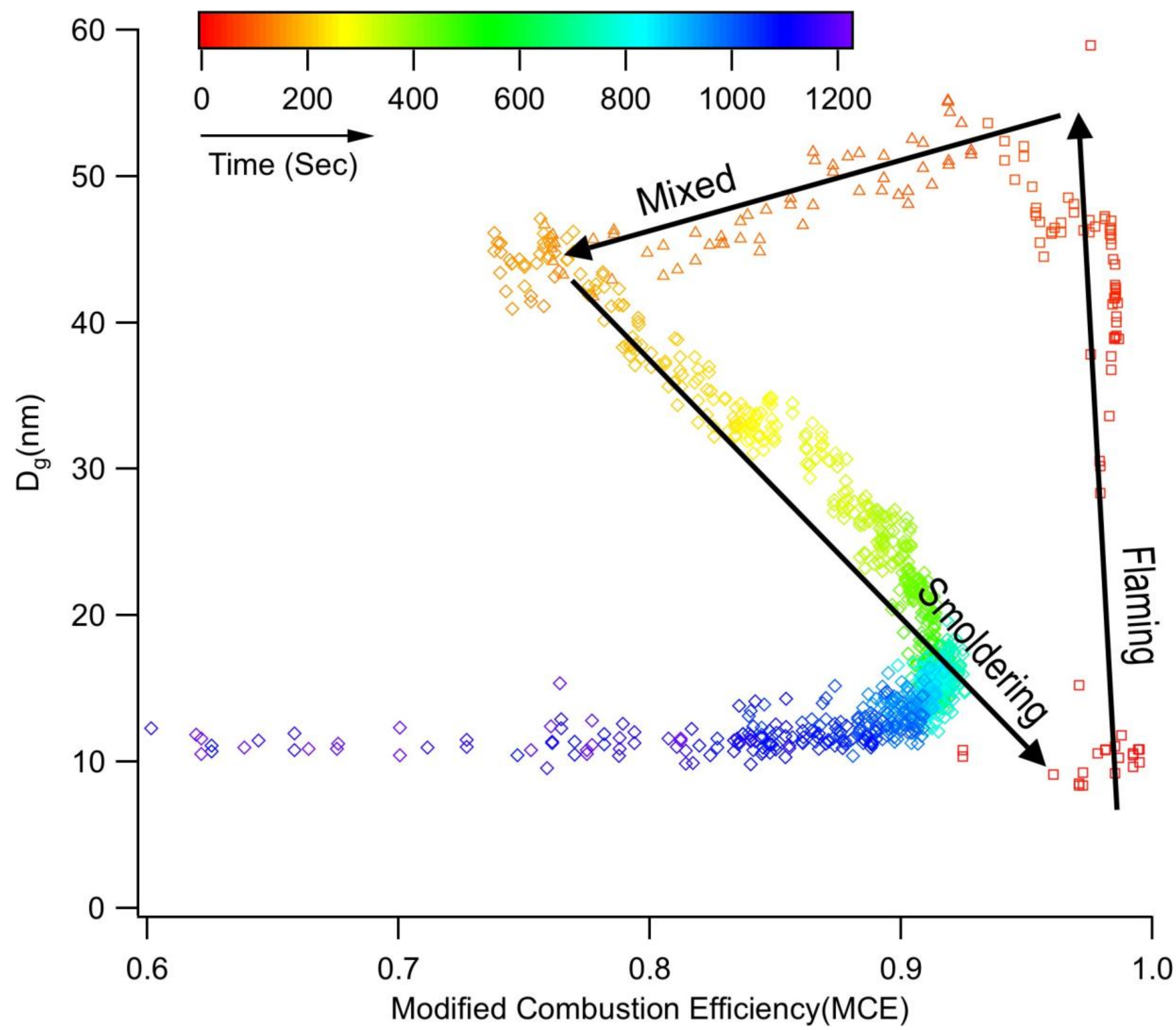
It seems to me that the spacing of diameters in the y-axes in Fig. 9 is very
narrow, such that any error introduced in the particle size distributions could mess up
the whole MCE vs. geometric mean diameter trend.

- ⇨ We disagree. Narrow spacing means more data point and longer period duration. For example you can see that data are dense in smoldering phase which has long duration. The Figure 9abc are obtained based on real time data of three specific burns from FMPS measurement. For clarification we drew new figures showing time course of the data. Please see below.

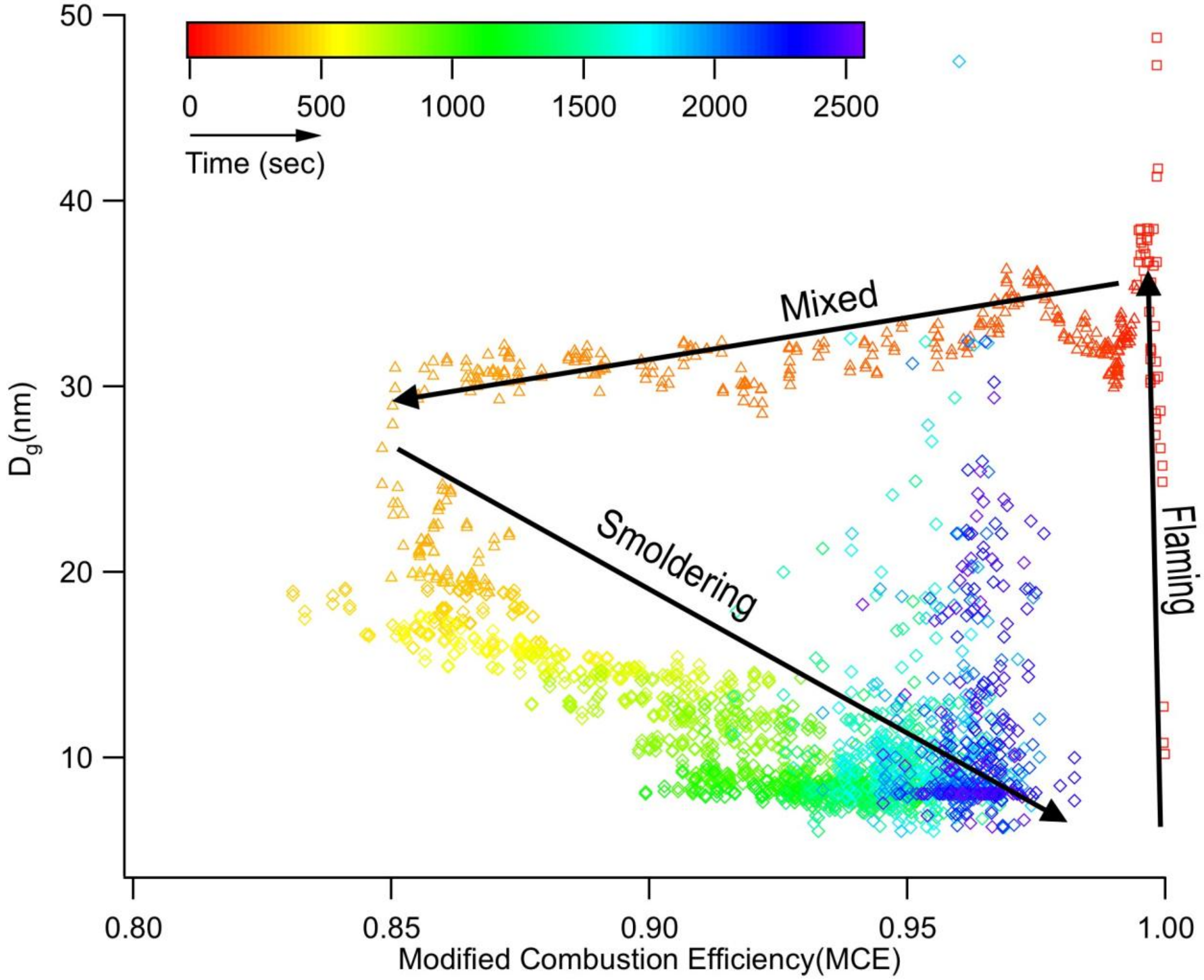


⇨

New Figure 9(a)



New Figure 9b



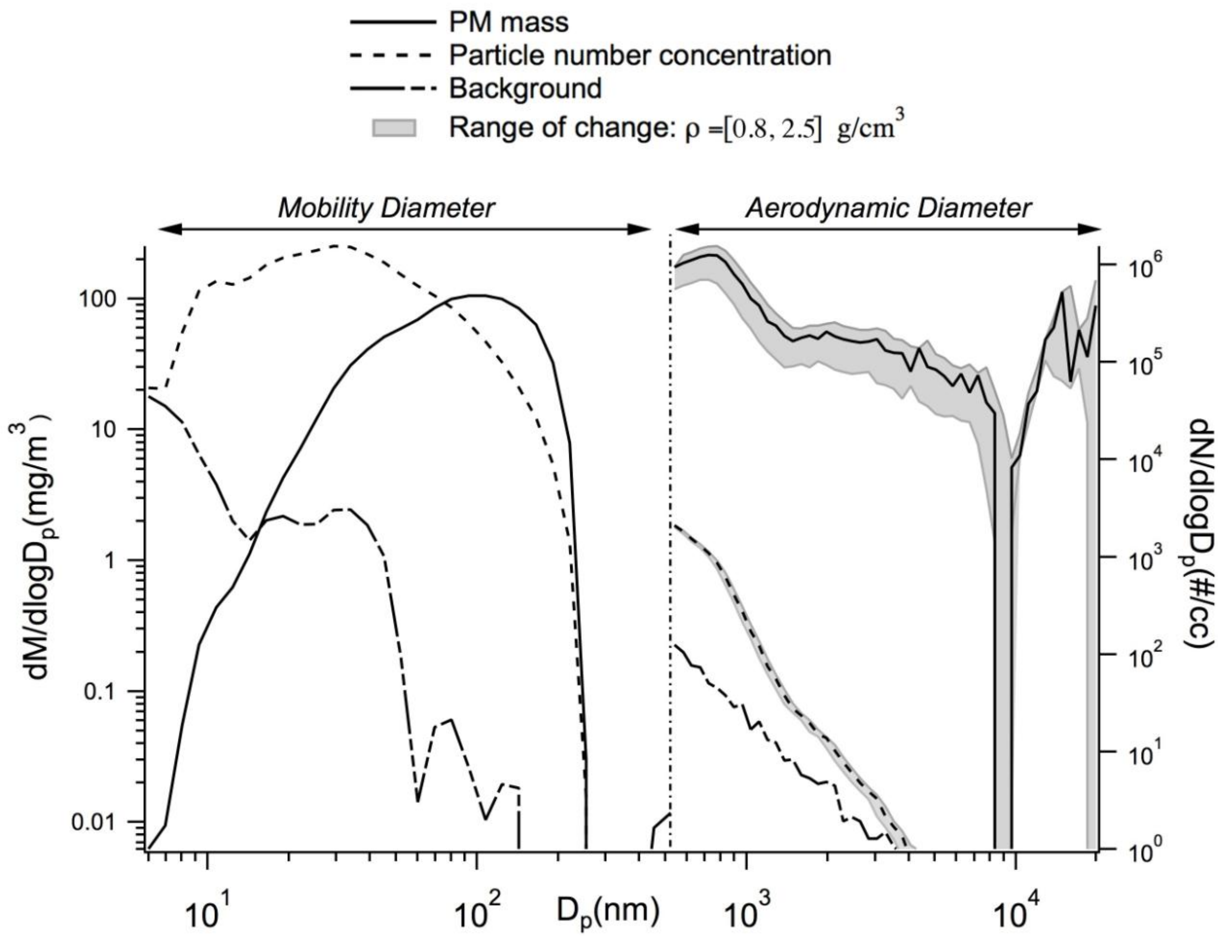
New Figure 9c

That is why I strongly suggest the authors to flesh out the details of their particle size distribution methodology and then claim the MCE vs. geometric mean diameter trend.

4) In Figure 6, the authors claim that the particles larger than 0.5 micron attribute to 30% of total volume measured by APS and FMPS. Again, this study involves measuring fractal-like particles from flaming phase, which implies that the size distribution will show significant differences in the size distributions measured by mobility and aerodynamic techniques. That said, I would want to see an error bar introduced in the "30%". During the course of this experiment, the authors never take into account of the effect of flow regime on aerodynamic diameter measurement. Significant corrections apply in the APS for large fractal-like particles with Reynolds number greater than 0.5, because in this case the drag is non-Stokesian. Secondly, relative humidity (RH) could also affect the measurements, and hence, information on RH could be very helpful. Thirdly, for fractal-like particles of same density, their mobility diameter is greater than their

aerodynamic diameter (refer to Baron, P. A., and Willeke, K. (2001). Gas and Particle Motion. In Aerosol Measurement:Principles, Techniques, and Applications, edited by P. A. Baron and K. Willeke. Wiley, New York). Therefore, it could be very well possible that the total volume be greater than 30%. I would like to see all these issues addressed in the manuscript.

- ⇨ We appreciate the reviewer on this comment. As mentioned above there is absence of fractal-like particles in the measurement range of APS. However it is correct that there should be uncertainty due to non-Stokesian effects. We assumed two extreme densities of 0.8 and 2.5 g/cc for the particles the APS measured in our study. That gave the range of 51 to 68 % for the mass APS measured compared to the total mass. Below is the new figure. We also fixed dM/dlogDp for the APS in Fig 6. The original figure had wrong conversion in the graph.
- ⇨



- ⇨ Please note that the dilution was applied for all online aerosol measurement. RH of dilution air was maintained at 16.5±4%.

C3560
Minor comments:
1) In the abstract, the abbreviations (like US, USDA, vs.) need to be spelled out.
=> Fixed

2) Section 3.1: How did the authors ensure that the burns were similar to actual or real burning condition as claimed?

- ⇨ Below paragraph will be added prior to Section 3.1

- ⇨ Bulk density of the fuel beds ranged from 5.8 to 14 kg/m3 and the packing ratio (defined as the ratio of fuel bed bulk density to fuel particle density) ranged from 0.010 to 0.024. These packing ratios are similar to those reported for laboratory fire spread experiments (Weise et al., 2005), but they are 1 to 2 orders of magnitude larger than packing ratios observed in the field which indicates the fuel beds were less porous than natural fuel beds. While our initial intent was to burn fuel beds similar to those found naturally, the fuel beds in the lab experiment were much drier with higher bulk densities than the fuel types they represent. We have recently completed several operational scale prescribed burns in the field in the same fuel types to determine the linkages between lab and field results.

3) Section 3.1 2nd para – Please elaborate on the other indicators (other than the MCE) the authors used to segregate the mode of combustion during each burn.
=> Fixed

4) Section 3.1 2nd para – Shouldn't it be "geometric mean diameter" instead of "geometric diameter". If no, then how did the authors calculate the geometric diameter for flaming phase particles (fractal-like)?
=> The reviewer is correct. It should be geometric mean diameter. We fixed it in the revision

5) Section 3.1 3rd para – "of" after "Analysis" needs to be added.
=> Fixed
6) Section 4: Typo – "partcle" in " Time averaged partcle concentrations▹▹▹."
=> Fixed
Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8595, 2010.

H. Wang, W. John (1987) Particle Density Correction for the Aerodynamic Particle Sizer, Aerosol Science and Technology, 6, 191-198

Yokelson, R., R. Susott, D. Ward, J. Reardon, and D. Griffith (1997), Emissions from smoldering combustion of biomass measured by open-path Fourier transform infrared spectroscopy, J. Geophys. Res., 102(D15), 18865-18877.

Goode, J., R. Yokelson, R. Susott, and D. Ward (1999), Trace gas emissions from laboratory biomass fires measured by open-path Fourier transform infrared spectroscopy: Fires in grass and surface fuels, J. Geophys. Res., 104(D17), 21237-21245.