Review of "Dilution impacts on smoke aging: Evidence in BBOP data"

By Anna L. Hodshire et al.

Anonymous Reviewer

Summary:

This manuscript derives empirical relationships for aerosol chemical age, number concentration, and mean mobility diameter which are useful for modeling the climate impact of biomass-burning aerosols. New insight from the BBOP measurement campaign is used to show that the rate of chemical aging of aerosol is affected by the "concentration" of the plume, presumably through decreasing photolytic rates with increased aerosol optical depth, and plume size, presumably because the core of larger plumes and are more protected from mixing with clean ambient air.

Ultimately, this is an interesting study but has a number of key assumptions that need to be further investigated or justified. Although the authors have weakened their language since the original draft due to comments from reviewers, they have not addressed the underlying concerns of the reviewers. This manuscript will require major revisions prior to considering publication.

General Comments:

1. In general, the language has improved but is still not very precise and there are grammatical issues.
2. There is a lack of details regarding SP-AMS measurements (as well as description of other instruments). There are only scattered references to the operating conditions/settings used for measuring OA in these plumes. Although the instrumentation is fully described in another manuscript, there is a minimum amount of information required for the SP-AMS measurement: vaporizers equipped and modes used (switching between modes, temperature of thermal vaporizer), calibration, description of CE determination, ToF mode (HR-ToF, C-ToF, V-mode, W-mode), MS sampling timing (Open, closed, PToF, ePToF, pulser period, etc)

   There also needs to be a description of the mass spectra analysis. What software was used. Are you reporting UMR or HR results? Is f60 based on m/z=60 or the specific ion $C_2H_4O_2^+$? How is gas phase subtracted? Assuming constant [CO_2] gas phase concentrations of 400 ppm?

   Since the vaporizer modes were switched (presumably intermittently) it is concerning how the authors choose to combine the data from the different modes. These modes measure inherently different components of the aerosol mass and fractionate the molecules in different ways. While Lee et al (2020) show that molecule fractionation in the different vaporizer modes is similar for the $C_2H_4O_2^+$ ion (used to calculate f60, although that is not stated by the authors), the fractionation is significantly different for the organic fraction of CO_2^+ (used to calculate f44) (see also Onasch et al 2012, Canagaratna et al 2015, etc).

3. Data are binned by physical age and further into edge-vs-core (such as shown in Figure 1). Each binned datum represents multiple measurements and therefore, in figure 1 and 2, should include error bars representing the variance of those measurements. This could be independent of measurement uncertainty, but would be better if it did include propagated instrument uncertainty.

4. A key assumption of the authors is that $\Delta OA_{initial}$, Δf60, and Δf44 can be used to identify dilution of the plume. However, these parameters are known to vary in primary emissions.

   In the manuscript, the authors support the assumption regarding $\Delta OA_{initial}$ with the measured Δf60 and Δf44. However, these parameters are all more likely related to variations in POA between fires and within a fire as fire conditions change. The Δf60 and Δf44 measurements are the only support the authors provide for their main conclusion.

   In the author's revisions, they have tried to further justify this assumption by making a flawed argument that their interpretation is only invalid if f60 and f44 covary with OA emissions. First this argument is flawed, as $\Delta f60_{initial}$ and $\Delta f44_{initial}$ are more reasonably attributed to differences in POA. Second, it has actually been shown that f60 and f44 of POA can covary with OA emission factors (see Corbin et al 2015; Ortega et al 2015; and Lee et al 2010).

5. Interpretation of Spearman's correlation coefficients and the strength of these coefficients is hindered by the authors choice to combine all data from all flights together for their regressions without normalization despite showing that both aerosol age and emission factors affect the parameter of interest (e.g. Δf_{60}). This multi-variate dependence is even stated several times by the authors. For example, with 1 exception, all transect sets predictably show that Δf_{60} decreases with physical age, but because different transect sets started with different f_{60} values the combined data set does not monotonically decrease with physical age and the regression results in a weak relationship ($R_{Spearman}$ = -0.26).

   This is an example of where the authors should rethink their analytical approach but have instead weakened the language of their results.

   There are several possibilities that the authors could consider. Continuing to use Δf_{60} as an example, the authors could:

- Normalizing the data to the initial measured value (e.g. Δf_{60}- Δf_{60}initial) prior to combining the data. This allows you to remove the processes driving variability in the initial Δf_{60} (essentially the emission factor) so you can isolate the effects of physical age on Δf_{60}.
- A multivariate analysis with predictors of $\Delta OA_{initial}$ and physical age. You could do this in any programming language, but using Excel as an example you would use the Data>Data_Analysis>regressions gui. It appears that you use this in your model in section 3.1 in equation 4. Since you did this, why do you even show the single-variable $R_{pearson}$ and $R_{spearman}$ values?
- Lastly, you could analyze each transect set separately to get a $R_{spearman}$ value of Δf_{60} versus physical age and then average those values together. If this approach is chosen, then averaging should weighted by the number of transects in the transect set. Also, you should use a jack-knife-like approach, repeating the averaging by systematically excluding 1-2 transects sets to see how dependent the results are on any individual set.

Currently, the analytical approach is inappropriate and therefore should not be used as support for their conclusions, regardless of the strength of language chosen to describe the empirical relationships.

7) SI section: The supplement has improved with more detail but is still lacking. More detail is needed describing the instrument set up, even if it is described fully in another paper.

The heterogeneous chemistry calculations needs a description of the calculation and justification for the methodology used. The only information provided is that it is a "simple calculation" and a list of what the parameters are.

Specific Comments:

L31: Here and elsewhere, are you calculating the mass mean mobility diameter or number mean mobility diameter?

L38: "…undergone more decreases in a marker for primary biomass burning organic aerosol." This is an awkward statement

L41-44: "Smoke from biomass burning… influencing… as well as the health of smoke-impacted communities". "Smoke-impacted" is redundant.

L45: Dilution is a process which is a central theme of the manuscript. It should have a proper description of what that process is. I suggest something like, "Dilution is the process where the plume mixes with clean background air, reducing concentration of fire emitted aerosols and gases". The current statement, "Dilution through entrainment…" is not explicit.

L50-52: Lacks explanation of why large plumes dilute slower. Since this is so important to the story, it should have a better description. Currently, it is just stated that they do. Something like "…cores of larger plumes are protected from dilution due the physical distance from background air…" Citations for this are Garofalo et al (2019) and Lee et al (2020).

L86-87: "… evaporation of vapors." Should be "evaporation of more volatile compounds."

L88: "; plumes with higher concentrations will undergo more coagulation…" Are you referring to number concentration, mass concentration, or both?

L101: "… differences in aerosol loading serve as a proxy for differences in dilution rates…" Do you mean rates or amount of dilution prior to first measurement? I have provided other comments in more detail regarding this assumption.

L103: "…given initial plume mass and physical age…" "mass" should be "OA mass concentration".

L115: More description needs to be provided regarding the settings for the SP-AMS.

Was it equipped with a tungsten thermal vaporizer? If so, what temperature was the thermal vaporizer set to? HR-ToF, C-ToF, L-ToF, quadrupole? ToF set to V-mode or W-mode? How was data analyzed? Are you reporting UMR or HR results? Is f60 the levoglucosan fraction (i.e. $fC_2H_4O_2$ as is discussed by Corbin et al 2015 and Lee et al 2020) or based on the UMR m/z=60 organic fragment after subtracting C5 contributions (also m/z=60, see Cubison et al 2011 and Lee et al 2010). What was the MS timing? Open vs closed timing? PToF or ePToF mode? Pulse period, sampling Hz? What m/z range was scanned?

L116: "PM1 aerosol masses" should be "aerosol mass concentration of PM1 (sub-micron particulates)…"

L119-121: How was collection efficiency determined?

L119-121: It looks like the laser vaporizer was switched between on and off. How frequently was the laser vaporizer switched? Is the data presented in this manuscript with the laser on, off, or both?

These measure inherently different attributes of PM1 that may not be directly comparable or combinable.

Also, because the laser vaporizer fractionates aerosols molecular species differently than the thermal vaporizer (Onasch et al 2012; Corbin et al 2014; Canagaratna et al 2015; Lee et al 2015; Lee et al 2020) single ions such as $C_2H_4O_2^+$ and $Org44^+$ (used to calculate f60 and f44) CANNOT be compared or combined between modes.

L121-L122: "We do not attempt to characterize whether the collection efficiency of the SP-AMS changes as the aerosol ages"

Collection efficiency has been observed to change by a factor of 2 or more as BB POA grows in size and becomes more spherical (See Middlebrook et al 2012, Willis et al 2014, Corbin et al 2015 (ACP), Massoli et al 2015, Collier et al 2018). This change in CE has been observed to bias particles of different morphology/composition differently between different vaporizer modes (laser + oven versus oven-only), specifically affecting the CE of the laser more than the CE of the thermal vaporizer.

L123: "…CE has been recently observed to decrease with aging within a laboratory study…" should be "… decrease with increasing chemical age induced by UV light exposure and OH-equivalent to 10 photochemical days…"

This change in CE is likely irrelevant to this manuscript since the physical age of aerosols described in this study is generally less than 3 hours while the cited study compares CE over the equivalent of 10 days. As mentioned in the previous comment, there are a range of studies that have shown increases in CE as particles grow in size (which presumably also increase with age in the near-field during the particle growth phase) which is more relevant to this study.

L 124: "… no consistent evidence of changing CE in field studies exist yet."

There are lots of studies which show changing CE in the field. For example, see Collier et al (2018), Massoli et al (2015), and Middlebrook et al (2012) for examples of changing CE in field studies. Also see Willis et al (2014) and Corbin et al (2015, ACP) which report the same phenomena in laboratory studies.

L124-125: "We use the f60 and f44 fractional components…"

Here and elsewhere, f60 and f44 are referred to as if they are chemical species instead of parameters describing mass fractions.

For f50, the relevant OA species group is anhydrous sugars with the dominant species being levoglucosan ($C_6H_{10}O_5$) (Lee et al 2010; Cubison et al 2012). This species is indirectly observed by the SP-AMS as the fragment $C_2H_4O_2$ (m/z=60), a fragment of the levoglucosan molecule after the OA is vaporized and the vapors are ionized by a 70 eV electron supply. Similarly, OA observed at m/z 44 is the CO_2 fragment of, primarily, OOA after subtracting the gas [CO2] mass.

L128-129: "The f44 fractional component (arising from primarily CO2+…)"

This is another example of imprecise language. Suggest, "f44, the OA fractional component observed by the SP-AMS as the ion fragment CO2+, is a proxy for …" The current wording suggests that f44 is a fraction of something else which isn't specified (i.e. f44/Org).

L129: extra semicolon

L130-132: "...of semivolatile <u>f60-containing</u> species and addition of oxidized <u>f44-containing</u>…" Another example of imprecise language using f60 and f44. The aerosols contain levoglucosan and anhydrous sugars, not f60, and OOA, not f44.

L135: "changes in O/C and H/C are also influenced by…"

These other processes also affect all of the other parameters discussed in this paper.

L140: "provided CO measurements" should be "measured CO concentration" or "measured CO mixing ratio".

L141: "An SPN1 radiometer provided total shortwave irradiance". The radiometer measured total shortwave irradiance, it did not provide or create the irradiance.

L143: "(Δx)" refers to the smoke contribution and should be placed after "species X from smoke" otherwise Δx is implied to refer to the background value of x. Also, the smoke doesn't contribute species X, it is comprised of species X. The fire emits/contributes X.

L145-146: This sentence needs to be rewritten. Suggest something like "Variability of the normalized emission ratio (Δx/ΔCO) along the lagrangian flight path implies production or removal of species X in the plume."

L147: "… average regional background for each species by using the lowest 10% of the CO data for…"

This statement reads as if you subtract a CO concentration from the number size distribution, OA, etc to get a background correction. It should say something like "… background values of X… were determined to correspond with time periods which displayed the lowest 10% of CO concentrations…"

L149: Should read "Mass concentrations of elemental O, H, and C were calculated…"

L152-153: It is not clear what you mean by this sentence.

L154: "inside and outside of the plume".

Does this refer to sampling time periods/locations corresponding lowest 10% of CO or <150 ppbv? Why would you change between these definitions of background?

L157-158: “We only consider data to be in-plume if the absolute CO>=150 ppbv, as comparisons of CO and the number concentration show that in-plume data has CO>150 ppbv and out of plume (background) data has CO < 150 ppbv.”

Why did you change the definition of background from time/location corresponding to lowest 10% of CO to <150 ppbv?

What do you mean by “comparisons of CO and the number concentrations”? What independent metric are you defining as “background” here? If number concentration is used to define time/location of background air, why not use that instead of CO <10% or CO < 150 ppbv? The logic here is circular or incomplete.

L 162: “concentrations” should be singular.

L163: Should be “mobility” diameters.

L164: “…as the bulk of <u>observed</u> newly formed particles <u>observed</u> fell below 40 nm” Grammar.

How do you identify “newly formed particles” independent of the particle size? This implies that you observed newly formed particles >40 nm.

L165-166: Grammar in sentence structure.

L187-188: “The centerline…” This sentence needs to be rewritten for clarity.

L190: “…and this physical age is assumed to be constant across the transect, as the crossings took between 50-500 seconds.”

While crossing the plume occurred in only a short time, were the transects always perfectly across flow? If not, then wouldn’t the aerosol at different sampling times along a transect have different physical ages with larger uncertainty than just 50-500 seconds?

L195: missing comma

L201-202: “thinnest (least CO-dense)… thickest (most CO-dense)…” Use either of the commonly accepted nomenclature of “CO mixing ratio” or “CO concentration”.

L207: missing a verb between “plumes” and “from”

L205-207: could the multiple peaks during a transect be explained by spatial variations in the plume structure, such as the core of the plume was higher in some areas than others causing the flight to dip below/above the core and then back into it?

L213: It is hard to tell what the variability in ΔBC/ΔCO are since they are plotted on a log scale. They appear to vary by an order of magnitude, i.e. not constant as the authors suggest.

L213: I pointed this out in my general comments. Each dot in figure 1 is a single value that represents multiple measurements in space and time. How well does the value of any single datum in this figure represent the range of data it is derived from? You need to have error bars to show that variability.

L215: “for each transect” should be “each transect set”.

L218-219: “…it is apparent that the 5-15 and 9-90 percentiles do show a separation…”

This statement cannot be verified or supported without some idea of error bars, either representing propagated measurement uncertainty, variability of binned data, or both.

L227-231: This is one of the key assumptions of the research, that the initial, background-corrected OA mass concentration can be used as a proxy for the degree of dilution of the plume. The authors provide no support for why $\Delta OA_{initial}$ would represent the degree of dilution even though this is the main storyline of their paper.

First, I do agree that the cores of larger plumes are likely protected from mixing with background air because of the distance between the core and the background air and it is well understood that some plume chemistry and mixing occurs very early after combustion+pyrolysis and prior to measurement. However, this needs to be presented differently. Start with the hypothesis that cores of larger plumes mix slower with background air and then use the observations to prove it by showing something like the rate of change in f60 and f44 as a function of physical age for plumes with different $\Delta OA_{initial}$.

The current presentation is problematic. Think of two hypothetical smoke plumes that are identical in terms of dilution, photolytic reaction rate etc and were measured at the same physical age, but the corresponding fires had different OA emission factors (say, flux of OA from fire B was twice that of fire A). Fire B would have ~2x the measured $\Delta OA_{initial}$ compared to Fire A. This would instead be interpreted by the authors as having half the dilution of the plume from fire B instead of twice the OA emission.

This demonstrates that the author’s assumption that $\Delta OA_{initial}$ is a proxy for plume dilution only makes sense if all fires measured emit OA at the same rate and concentration.

L228-229: “(as presumably larger, more intensely burning fires will have larger mass fluxes than smaller…)”.

This assumption is false.

Larger and more intense fires do not necessarily correspond to higher emission rates. Emissions of OA depend on a number of factors other than fire intensity (I assume you mean temperature). Hotter, more intense fires (i.e. flaming stage) can burn more efficiently and actually emit less OA than cooler, smoldering fires (Akagi et al 2011; McMeeking et al 2009; May et al 2014). Corbin et al (2015) found that in laboratory burn experiments the vast majority of OA emissions occurred in the "starting phase" before the logs fully caught fire.

L243: missing a comma between items in the list.

L252: delete either "systematically" or "sequentially"

L251-257: See general comments regarding data analysis. The authors choose to combine all data together to determine the effects of aging and dilution on plume characteristics. However, they do not normalize the data in anyway or try another technique to separate these two effects. Normalizing a parameter to the first measured value (say f60) acknowledges that there are differences in f60 between plumes (maybe related to $\Delta OA_{initial}$) and would allow for analysis of temporal trends after emission. One result maybe that the photolytic age of the aerosol mass (as measured by f60 or f44) is slower plumes with higher $\Delta OA_{initial}$, i.e. there is less of a change/unit time of f60 or f44 or $\Delta N/\Delta CO$, etc.

Currently, the analysis is a regression comparing apples and oranges and the results are not meaningful.

L262-266: Include citations to Cubison et al (2011), Garofalo et al (2019), Forrister et al (2015), Lee et al (2020) for constant $\Delta OA/\Delta CO$ as plumes age.

L270-270: Containing text "…estimates of heterogeneous mass losses indicate that <u>after three hours</u> of aging for a range of OH concentrations and reactive uptake coefficients, over 90% of aerosol mass is anticipated to remain…"

This is the basis for which the authors interpret changing f44, f60 as relating to coagulation. However, this statement is only relevant to particle evolution after ~3 hours while nearly all of the observations occur within a physical age of 3 hours.

L281: "with more concentrated plumes". Be more specific by what you mean by more concentrated. Do you mean less diluted, higher mass concentrations of OA, higher CO mixing ratios?

L282-284: "(2) Differences in $\Delta f60$ and $\Delta f44$ for the nearest-to-source measurements indicate that evaporation and/or chemistry likely occurred before the time of these first measurements…"

It is well documented that the f60 and f44 of POA varies between fires (Cubison et al 2011; Jolleys et al 2015; Ortega et al 2013; McClure et al 2020). Since differences of POA emissions can explain variability in Δf60 and Δf44 for the nearest-to-source measurements, variability of these parameters can NOT be used as evidence of chemistry/evaporation in the smoke plume without knowing the actual f60 and f44 values of fresh POA.

L282-284 "…(assuming that emitted Δf60 and Δf44 do not correlate with $\Delta OA_{initial}$; there is currently no evidence for this alternative hypothesis)."

There is actually a lot of evidence that f60 and f44 can correlate with OA emissions. In laboratory studies, the evolution of emissions as fires progress from starting-to-flaming-to-smoldering has shown that levoglucosan emissions occur primarily at the starting phase by combustion of hemicellulose material which is also when the majority of OA emissions occur (Corbin et al 2015). Ortega et al (2015) and Lee et al (2010) also observed increased values of f60 in lab burns with higher OA emission factors. These laboratory studies support observations of smoke in ambient troposphere (e.g. Lee et al 2010; Aiken et al 2009; Lee et al 2020).

As you note, it is hard to measure the f60 of POA in ambient smoke. However, the lifetime of levoglucosan in the free troposphere is much longer than the age of aerosol in this study (<3 hours) which is probably why there is only a weak trend to lower f60 values with increasing physical age. So, your measurements of Δf60 should be fairly representative of POA and your study (and your first point on lines 280-282 and repeated on line 319) are evidence that f60 is correlated with OA emission factors in wildfires.

L284: "Amounts" should be singular.

L291: Add citation Jolleys et al (2015).

L313-314: delete "tends to be fairly constant or slightly decreasing with physical age and". Saying that it is poorly correlated is enough.

L319-320: Evaporation does not happen from dilution. Evaporation will happen if the air is undersaturated (less than predicted vapor pressure of species X compared to equilibrium predicted by Henry's law). Here and elsewhere, please don't say that dilution causes evaporation, instead that dilution promotes evaporation.

L334: "NME is <u>more variable</u>…" Do you mean larger or higher? The NME is more variable between parameters, but that is meaningless.

L336: too many open brackets.

L337: What do you mean by "biomass burning modeling"? Are you referring to models of BB emissions, aerosol aging, fire spread?

L343-345: Since you present a mulit-variate analysis here, what was the point of the past several pages discussing single variable correlation coefficients? Especially after you show that

"Both physical age and $\Delta OA_{initial}$ appear to influence Δf_{60}, Δf_{44}, and $\Delta O/\Delta C$…" (Line 319 and a similar statement on L280-282).

L360-363: This statement needs more explanation and needs citations. Why would you expect plume regions with higher $\Delta OA_{initial}$ to have lower normalized number concentrations?

To a first order, I would expect the opposite. That higher number concentrations would be observed with higher $\Delta OA_{initial}$ because the OA vapor pressure is higher and this promotes new particle nucleation because vapors are more likely to collide with other vapor molecules to nucleate than with existing particles to condense on (Lim et al 2019; and work from Neil Donahue's group).

L371-372: Decreasing normalized number concentrations are ascribed to coagulation. This contradicts the model that changes in f60 and f44 are due to evaporation of solid particulate balanced by condensation of more oxidized-OA described on line 277-279.

L381-382: Awkward and redundant sentence.

L384-393: Discussion of nucleation-mode particles seems out of place here. Maybe move towards beginning of section 3.

Nucleation-mode particles are defined as 20-40nm. This needs a citation.

Earlier, a statement was made "bulk of observed newly formed particles observed fell below 40 nm" which implies that a fraction of newly formed particles were larger 40 nm. Was that a mis-statement? This would contradict your definition that nucleation-mode particles are 20-40 nm?

L388: "one day" should be one transect or transect set.

L405-406: Awkward or redundant sentence.

L408-409: "indicate that evaporation and/or chemistry has likely occurred before the time of initial measurement…" See previous comments questioning validity of this statement.

L437: Format of some citations need to be cleaned up.

Figure 1:

- See comments regarding adding error bars to show data variability of bin. There are a few points that are very different from the rest of the data set (such as in the $\Delta OA/\Delta CO$ and $\Delta N/\Delta CO$ datasets) which makes me think the single value representing the bin is inadequate.
- Please change $\Delta BC/\Delta CO$ to a linear scale

- Your values of f60 are pretty low for fresh BBOA. I am wondering if this is an issue with the SP-AMS settings or how the data was handled.
- Need units for Dp axis, “[nm]”.

Figure 2:

- If you insist on combining all of the data together for a single regression, than you should not be drawing lines between points. Instead this should be a scatter plot with markers.
- Legend is inconsistent with figure 1. Either use “edge” or $5\%<\Delta[CO]<15\%$.
- Use of “[CO]” is inconsistent with text.
- Need units for Dp axis
- Caption says that panels (d) and (g) have log axis but are plotted on linear axis. Panels (a) and (f) are plotted on log axis (also in corresponding figures in SI).
- Font of “Dp” in caption is different than rest of fonts.
- I think you should also provide a scatter plot of the first measurement of these parameters as a function $\Delta OA_{initial}$.
- Needs error bars

Figure 3:

- Spearman’s correlations are not needed here.

Supplemental Information:
“…electrical mobility as in SMPS…” Should be “…similar to the operating principle of the SMPS…”

“…when size distribution suggests that particles smaller than 10 nm contribute negligibly…” Neither the FIMS or CPC 3010 are efficient at counting <10nm Dm particles, so why would the existence of those particles cause differences between the two instruments?

“The SPAMS is thoroughly detailed in Kleinman et al. (2020)…” This still needs to be described here. At least summarizing the operating conditions of the SP-AMS. 1

“An SPN1 radiometer provided total shortwave irradiance…” It probably measured total shortwave irradiance, not “provided” or “created” the irradiance. This instrument needs to be described more. Maybe what exactly the measurements are and what they represent.

“… following parameters assumed for the calculation” missing a verb.

“Heterogeneous chemistry calculations:” There is no citation to justify the calculation. Is this a common methodology used? Has this methodology passed peer review?

Fig S1: Colorbar label is missing an “]”

Fig S7-S11: Why are x-axis on the top and bottom panels different scales?

Fig S13: should be moved to just before figure S19

Fig S19-S22: Caption mis-identifies which panels use a log scale.

Fig S24: Could you also plot the [CO] of each transect similar to S24-S26. I want to see that the absolute concentration is higher in the center of the plume than the edges and that the [CO] of the core decreases in each successive transect of the set to show dilution.

Fig S27: Need the 1:1, 1:2, and 0.5:1 line representing constant lines of oxidation. If it is arbitrary where the intersection of these lines is placed (as you cite from Heald et al 2010), then have an arbitrary intersection near the average of your data. The importance is the trends in H:C vs O:C. Alternatively, remove this figure.

References:

- Aiken, A., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L. (2009). Mexico City Aerosol Analysis During MILAGRO Using High Resolution Aerosol Mass Spectrometry at the Urban Supersite (T0)—Part 1: Fine Particle Composition and Organic Source Apportionment. Atmos. Chem. Phys. 9:6633–6653.

- Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039–4072, https://doi.org/10.5194/acp-11-4039-2011, 2011.

- Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., et al. (2015). Elemental ratio measurements of organic compounds using aerosol mass spectrometry: Characterization, improved calibration, and implications. Atmospheric Chemistry and Physics, 15(1), 253–272. https://doi.org/10.5194/acp-15-253-2015

- Collier, S., Williams, L. R., Onasch, T. B., Cappa, C. D., Zhang, X., Russell, L. M., et al. (2018). Influence of emissions and aqueous processing on particles containing black carbon in a polluted urban environment: Insights from a soot particle-aerosol mass spectrometer. Journal of Geophysical Research: Atmospheres, 123, 6648–6666. https://doi.org/10.1002/2017JD027851

- Corbin, J. C., Sierau, B., Gysel, M., Laborde, M., Keller, A., Kim, J., et al. (2014). Mass spectrometry of refractory black carbon particles from six sources: Carbon-cluster and oxygenated ions. Atmospheric Chemistry and Physics, 14(5), 2591–2603. https://doi.org/10.5194/acp-14- 2591-2014

- Corbin, J. C., Lohmann, U., Sierau, B., Keller, A., Burtscher, H., and Mensah, A. A.: Black carbon surface oxidation and organic composition of beech-wood soot aerosols, Atmos. Chem. Phys., 15, 11885–11907, https://doi.org/10.5194/acp-15-11885-2015, 2015.

- Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D., Lechner, M. J., et al. (2011). Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies. Atmospheric Chemistry and Physics, 11(23), 12,049–12,064. https://doi.org/ 10.5194/acp-11-12049-2011

- Forrister, H., et al. (2015), Evolution of brown carbon in wildfire plumes, Geophys. Res. Lett., 42, 4623–4630, doi:10.1002/2015GL063897.

- Jolleys, M. D., Coe, H., McFiggans, G., Taylor, J. W., O'Shea, S. J., Le Breton, M., Bauguitte, S. J.-B., Moller, S., Di Carlo, P., Aruffo, E., Palmer, P. I., Lee, J. D., Percival, C. J., and Gallagher, M. W.: Properties and evolution of biomass burning organic aerosol from Canadian boreal forest fires, Atmos. Chem. Phys., 15, 3077–3095, https://doi.org/10.5194/acp-15-3077-2015, 2015

- Lauren A. Garofalo, Matson A. Pothier, Ezra J. T. Levin, Teresa Campos, Sonia M. Kreidenweis, and Delphine K. Farmer (2019): Emission and Evolution of Submicron Organic Aerosol in Smoke from Wildfires in the Western United States, *ACS Earth and Space Chemistry 3* (7), 1237-1247. DOI: 10.1021/acsearthspacechem.9b00125

- Taehyoung Lee, Amy P. Sullivan, Laura Mack, Jose L. Jimenez, Sonia M. Kreidenweis, Timothy B. Onasch, Douglas R. Worsnop, William Malm, Cyle E. Wold, Wei Min Hao & Jeffrey L. Collett Jr. (2010) Chemical Smoke Marker Emissions During Flaming and Smoldering Phases of Laboratory Open Burning of Wildland Fuels, Aerosol Science and Technology, 44:9, i-v, DOI: 10.1080/02786826.2010.499884

- Lee, A. K. Y., Willis, M. D., Healy, R. M., Onasch, T. B., & Abbatt, J. P. D. (2015). Mixing state of carbonaceous aerosol in an urban environment: Single particle characterization using the soot particle aerosol mass spectrometer (SP-AMS). Atmospheric Chemistry and Physics, 15(4), 1823–1841. https://doi.org/10.5194/acp-15-1823-2015

- Lee, J. E., Dubey, M. K., Aiken, A. C., Chylek, P., & Carrico, C. M. (2020). Optical and chemical analysis of absorption enhancement by mixed carbonaceous aerosols in the 2019 Woodbury, AZ, fire plume. Journal of Geophysical Research: Atmospheres, 125, e2020JD032399. https://doi.org/ 10.1029/2020JD032399

- Lim, C. Y., Hagan, D. H., Coggon, M. M., Koss, A. R., Sekimoto, K., de Gouw, J., Warneke, C., Cappa, C. D., and Kroll, J. H.: Secondary organic aerosol formation from the laboratory oxidation of biomass burning emissions, Atmos. Chem. Phys., 19, 12797–12809, https://doi.org/10.5194/acp-19-12797-2019, 2019.

- Massoli, P., et al. (2015), Characterization of black carbon-containing particles from soot particle aerosol mass spectrometer measurements on the R/V Atlantis during CalNex 2010, J. Geophys. Res. Atmos., 120, 2575–2593, doi:10.1002/2014JD022834.

- May, A. A., et al. (2014), Aerosol emissions from prescribed fires in the United States: A synthesis of laboratory and aircraft measurements, J. Geophys. Res. Atmos., 119, 11,826–11,849, doi:10.1002/ 2014JD021848.

- McMeeking, G. R., et al. (2009), Emissions of trace gases and aerosols during the open combustion of biomassin the laboratory, J. Geophys. Res., 114, D19210, doi:10.1029/2009JD011836.

- Ann M. Middlebrook, Roya Bahreini, Jose L. Jimenez & Manjula R. Canagaratna (2012) Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, Aerosol Science and Technology, 46:3, 258-271, DOI: 10.1080/02786826.2011.620041

- Onasch, T. B., Fortner, E. C., Trimborn, A. M., Lambe, A. T., Tiwari, A. J., Marr, L. C., et al. (2015). Investigations of SP-AMS carbon ion distributions as a function of refractory black carbon particle type. Aerosol Science and Technology, 49(6), 409–422. https://doi.org/ 10.1080/02786826.2015.1039959

- Ortega, A. M., Day, D. A., Cubison, M. J., Brune, W. H., Bon, D., De Gouw, J. A., & Jimenez, J. L. (2013). Secondary organic aerosol for- mation and primary organic aerosol oxidation from biomass-burning smoke in a flow reactor during FLAME-3. Atmospheric Chemistry and Physics, 13(22), 11,551–11,571. https://doi.org/10.5194/acp-13-11551-2013

- Willis, M. D., Lee, A. K. Y., Onasch, T. B., Fortner, E. C., Williams, L. R., Lambe, A. T., Worsnop, D. R., and Abbatt, J. P. D.: Collection efficiency of the soot-particle aerosol mass spectrometer (SP-AMS) for internally mixed particulate black carbon, Atmos. Meas. Tech., 7, 4507–4516, https://doi.org/10.5194/amt-7-4507-2014, 2014.