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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observations and analysis of organic aerosol evolution in some prescribed fire smoke plumes" *by* A. A. May et al.

W. Morgan (Referee)

will.morgan@manchester.ac.uk

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The manuscript is well written and adds to the relatively scarce number of observations of fresh smoke plumes and their subsequent evolution. This is a timely and helpful addition to the literature, given the somewhat conflicting nature of previous observations. The manuscript adds insights into the potential drivers of the transformation of organic aerosol in the initial stages of smoke plume evolution, which is appropriate for ACP.

The manuscript is suitable for publication once some issues with the data analysis have been cleared up and some minor issues have been addressed.

P1955, L19-20: The authors suggest that 'increases in f44 are typically interpreted as indicating chemical production of SOA' – I would say that typically, increases in f44 are

thought to 'typically' indicate aging of SOA, rather than formation. It can be indicative of formation. I would suggest clarifying the text here.

P1957, L21-26: Is it really necessary to say that this is the first study to use that combination of instruments, especially when most of them have little or no use in this manuscript? If it is necessary, a sentence to illustrate why would be useful. At the moment it just seems like unnecessary boasting.

Section 2.1: It would be useful to include the typical altitudes for the sampling e.g. at what altitude was the close-to-source sampling conducted and what altitude was the downwind sampling conducted? Approximately how old was the initial smoke that was sampled close-to-source? This is useful context for both this manuscript and future studies that will likely cite this work.

P1960, L6: What does 'adjusted' mean in this context? Was the data simply averaged to the AMS time base or were the time series shifted to account for differences in inlet and/or instrument lag times? If so, how was this done?

Section 2.2.1: The uncertainties relating to the AMS collection efficiency (CE) should be expanded on here and a discussion of how they may affect the latter analysis should be included.

It is not clear how appropriate the Middlebrook et al. CE calculator is for aerosol that is dominated by organic material (such as biomass burning). As the authors are aware, there is a large range of AMS CE values (approx. 0.5 to 1.0) reported in the literature for biomass burning aerosol, which can introduce an additional uncertainty of a factor of two. Was an external measurement available during SCREAM that could be used to validate the AMS CE calculation? Furthermore, do the authors have any insights from their prior biomass burning datasets that may help to validate the use of this procedure?

The authors refer to May et al. (2014) for further details regarding the AMS data quality assurance for the inorganic species and I see that they have made fairly typical

adjustments to the fragmentation table for nitrate and sulphate ions. Given that the Middlebrook et al. CE calculator evaluates the CE depending on the contribution of the inorganic species, I wonder how much impact any composition changes in the plumes downwind will impact on the CE? Does nitrate form in the plumes downwind and does this impact the CE? Does the acidity of the aerosol evolve downwind (this requires careful and uncertain analysis of the ammonium contribution also, which is challenging for biomass burning aerosol with the AMS)? Discussion of these issues is required and how the uncertainty in the CE may impact the reported significance of the observed trends downwind should be included in the revised manuscript.

P1961, L1: Clarify that with no particle time-of-flight data being collected, no size-resolved information is available from the AMS.

P1961, L3-L11: As noted by Referee #1, this is misleading regarding the CO2 correction for the AMS.

Section 2.2.4: As noted by Referee #1, is this section necessary?

P1966, L15: Is it appropriate to call these 'Lagrangian'? This assumes that the fire characteristics and emissions are fixed over the time span between sampling the initial smoke and its subsequent evolution downwind. Do the measurements support this (the manuscript suggests not on P1966, L25)? Just flying along the plume does not guarantee this given that the speed of the aircraft and the speed by which the smoke is transported is not synchronised. I would suggest changing the terminology here or better defending this classification.

P1967, L25-28: This framework was also demonstrated in a partner paper to the Ng et al. (2010) paper in Morgan et al. (2010), which should be referenced here. The reference is included below.

Morgan, W. T., Allan, J. D., Bower, K. N., Highwood, E. J., Liu, D., McMeeking, G. R., ... Coe, H. (2010). Airborne measurements of the spatial distribution of aerosol

chemical composition across Europe and evolution of the organic fraction. Atmospheric Chemistry and Physics, 10(8), 4065–4083. http://doi.org/10.5194/acp-10-4065-2010.

P1968, L2: This is a somewhat confusing definition of f44 for those unfamiliar with the AMS, as it suggests that C44 is the mass concentration of particulate CO2+. While this is correct as far as the AMS fragmentation pattern is concerned, the CO2+ particulate signal is thought to arise due to decarboxylation on the vaporiser surface, rather than carbon dioxide being present in the actual aerosol sample. This should be clarified in the revised manuscript.

Figure 1: I suggest using a more colour-blind friendly scale on these flight tracks. Panel a) is particularly difficult to judge the differences. Color Brewer (http://colorbrewer2.org/) is a very useful resource for colour-blind friendly scales.

Figure 2: There is a seemingly large variation in the emission factors for CO and CO2 in these figures. What do the authors attribute this to and how does it affect the interpretation of the results?

Figure 6: There appears to be significant overlap between the near-source and downwind data for FJ 9b and Francis Marion fires. What do the authors attribute this to? As noted previously, it would be useful to include more details regarding the near-source samples.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 1953, 2015.