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Interactive comment on "The evolution of biomass-burning aerosol size distributions due to coagulation: dependence on fire and meteorological details and parameterization" *by* K. M. Sakamoto et al.

Anonymous Referee #1

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This paper studies the effect of coagulation on particle diameter (Dm) and geometric standard deviation (sigma) in biomass burning plumes using a large-eddy simulation model with an online aerosol microphysical module. The topic is timely and the text is well written; however, there are some issues that need to be clarified before this manuscript can be accepted. In my opinion this paper presents a valuable base case for assessing the role of coagulation and condensation in future biomass burning studies.

Major comments

I have two major concerns regarding this study: the lack of organic aerosol (OA) chemistry and that while interpreting the results little attention is placed on plume dilution and its effect on coagulation.

Running the simulations as "coagulation-only" limits the usability of the results. For instance, new particle formation has been observed in biomass burning plumes (Hennigan et al., 2012; Vakkari et al., 2014), as well as up to a factor of 4 mass increase during the first few hours (Vakkari et al., 2014). Also a recent study by Konovalov et al. (2015) suggests that accounting for OA volatility can improve model performance significantly, although over a much longer time scale than what is considered here. Therefore, without OA evaporation and condensation, can the parameterisations in this paper be a good starting point for global and regional scale models (c.f. line 37)?

On lines 453-455 it is concluded that SOA formation within the plume has a minor effect compared to coagulation. However, this is based on the assumption that SOA formation does not alter coagulation rate or sigma (lines 388-389). Could you elaborate on the conditions when these assumptions hold? For instance Pierce and Adams (2009) showed that secondary aerosol formation rate is one of the key parameters affecting how large fraction of small particles can grow up to CCN-sizes in new particle formation.

My second major concern is related to the effect of plume dilution on the coagulation rate. Coagulation depends strongly on aerosol particle number concentration (as stated on lines 145-146). However, the observed changes in Dm and sigma are not discussed in terms of concentration, but only with respect to the input parameters and a rather arbitrary dM/dx (aerosol mass in an infinitesimally thin slice of air perpendicular to wind direction). The effect of dilution on coagulation is mentioned only briefly (e.g. lines 324-326), though Figure 4 shows that in most simulations the Dm and sigma change rapidly near emission, but very slowly later on. Is this decrease in the rate of change in Dm and sigma due to plume dilution and subsequent slowing of coagulation rate?

How does Figure 4 look like if you colour it with concentration instead of dM/dx, or plot Dm and sigma against concentration? The dM/dx takes into account only dilution along the wind direction, not dilution due to vertical or cross-wind mixing.

Can you identify a range (time and space), where coagulation can cause significant changes in the size distribution and after which the plumes become so diluted that coagulation slows down? How would this turning point depend on the initial concentration (emissions) and the meteorological conditions (turbulent mixing) during transport?

The background aerosol is assumed to be negligible compared to the plume and is set to zero (lines 176-179). However, in ambient air measurements this assumption cannot be made – see e.g. Yokelson et al. (2009). Have you verified that your plumes are so concentrated even after 200km transport that this assumption still holds? When will coagulation rate with the background aerosol become similar to coagulation within the biomass burning mode?

How is turbulent mixing handled in the simulations? Table 1 (page 23) lists "Mixing depth of aerosol layer" as an input parameter, yet on line 214 "mixing depth" is calculated from the simulated vertical profile of aerosol mass. Is this related to mixed layer height (e.g. height of convective planetary boundary layer)?

If Figure 3 is a representative sample of the simulations it seems as if majority of the plumes are not in the mixed layer but above it, as they do not reach the surface. Again, I would expect the turbulent mixing in convective PBL (or the lack of convective mixing in free troposphere or the residual layer) to have a significant effect on plume dilution and therefore the coagulation rate. Is it so?

Minor comments

Line 69-71 There are some more recent studies which you might want to look up. For instance Akagi et al. (2012), Hennigan et al. (2012), Ortega et al. (2013), Vakkari et al. (2014), Jolleys et al. (2015), Konovalov et al. (2015) and May et al. (2015) come to

my mind.

Line 71 "This SOA condenses onto existing particles causing growth of the aerosol size distribution." Please reconsider this statement as there are observations of new particle formation in biomass burning plumes (Hennigan et al., 2012; Vakkari et al., 2014).

Line 153 "Mixing depth had a range of 150-2500 m" but Table 1 (page 23) gives "Mixing depth" limits as 120 m and 2500 m. Which one is it?

Line 169-170 "The algorithm simulated the size distribution across 15 logarithmically-spaced size bins spanning 3 nm-10 μm." This leaves quite few bins for the size range of interest. Can coarse size resolution become an issue for the coagulation calculation?

Line 191-192 "We ran 100 SAM-TOMAS simulations at 500 m x 500 m horizontal resolution (total horizontal extent = 100 km)," but Figure 4 x-axis extends to > 200 km. I assume these are the same data because on lines 244-245 it is stated that "Figure 4 shows the Dpm (panels a and c) and σ (panels b and d) as a function of distance for each of the 100 SAM-TOMAS simulations used to train the emulator (Sect. 3.2)." What was the horizontal extent?

Line 382-385 Also for this statement some more recent references could be considered.

Line 824, Figure 3 Please provide a legend for the lines (indicating input and meteorological parameters).

Line 871, Figure 4 There are so many overlying lines that it is getting difficult to read. Please consider if you can clarify it.

References

Akagi, S. K., Craven, J. S., Taylor, J. W., McMeeking, G. R., Yokelson, R. J., Burling, I. R., Urbanski, S. P., Wold, C. E., Seinfeld, J. H., Coe, H., Alvarado, M. J. and Weise,

D. R.: Evolution of trace gases and particles emitted by a chaparral fire in California, Atmos. Chem. Phys., 12(3), 1397–1421, doi:10.5194/acp-12-1397-2012, 2012.

Hennigan, C. J., Westervelt, D. M., Riipinen, I., Engelhart, G. J., Lee, T., Collett, J. L., Pandis, S. N., Adams, P. J. and Robinson, A. L.: New particle formation and growth in biomass burning plumes: An important source of cloud condensation nuclei, Geophys. Res. Lett., 39(9), L09805, doi:10.1029/2012GL050930, 2012.

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(6), 3077–3095, doi:10.5194/acp-15-3077-2015, 2015.

Konovalov, I. B., Beekmann, M., Berezin, E. V., Petetin, H., Mielonen, T., Kuznetsova, I. N. and Andreae, M. O.: The role of semi-volatile organic compounds in the mesoscale evolution of biomass burning aerosol: a modeling case study of the 2010 mega-fire event in Russia, Atmos. Chem. Phys., 15(23), 13269–13297, doi:10.5194/acp-15-13269-2015, 2015.

May, A. A., Lee, T., McMeeking, G. R., Akagi, S., Sullivan, A. P., Urbanski, S., Yokelson, R. J. and Kreidenweis, S. M.: Observations and analysis of organic aerosol evolution in some prescribed fire smoke plumes, Atmos. Chem. Phys., 15(11), 6323–6335, doi:10.5194/acp-15-6323-2015, 2015.

Ortega, A. M., Day, D. A., Cubison, M. J., Brune, W. H., Bon, D., de Gouw, J. A. and Jimenez, J. L.: Secondary organic aerosol formation and primary organic aerosol oxidation from biomass-burning smoke in a flow reactor during FLAME-3, Atmos. Chem. Phys., 13(22), 11551–11571, doi:10.5194/acp-13-11551-2013, 2013.

Pierce, J. R. and Adams, P. J.: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, Atmos. Chem. Phys., 9(4), 1339–1356,

doi:10.5194/acp-9-1339-2009, 2009.

Vakkari, V., Kerminen, V.-M., Beukes, J. P., Tiitta, P., van Zyl, P. G., Josipovic, M., Venter, A. D., Jaars, K., Worsnop, D. R., Kulmala, M. and Laakso, L.: Rapid changes in biomass burning aerosols by atmospheric oxidation, Geophys. Res. Lett., 41(7), 2644–2651, doi:10.1002/2014GL059396, 2014.

Yokelson, R. J., Crounse, J. D., DeCarlo, P. F., Karl, T., Urbanski, S., Atlas, E., Campos, T., Shinozuka, Y., Kapustin, V., Clarke, A. D., Weinheimer, A., Knapp, D. J., Montzka, D. D., Holloway, J., Weibring, P., Flocke, F., Zheng, W., Toohey, D., Wennberg, P. O., Wiedinmyer, C., Mauldin, L., Fried, A., Richter, D., Walega, J., Jimenez, J. L., Adachi, K., Buseck, P. R., Hall, S. R. and Shetter, R.: Emissions from biomass burning in the Yucatan, Atmos. Chem. Phys., 9(15), 5785–5812, doi:10.5194/acp-9-5785-2009, 2009.

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