Bian et al. investigate processes governing biomass-burning organic aerosol evolution in smog chamber and ambient plumes with the use of an aerosol microphysics model. An extensive set of sensitivity analysis is performed to evaluate the role of vapor wall loss, file size, dilution, and atmospheric stability in the SOA formation and evolution. Overall, the topic is very important and the manuscript is well written. One concern is the extent to which the simulations could adequately represent the actual measurements in chamber- and/or field-derived biomass-burning plumes. Specific comments are listed below that I would like the authors to address (consider) before publication in ACP.

General:

The authors found that accounting for vapor wall losses leads to 2-3 times increases in the total SOA production in chamber experiments. This conclusion, however, depends on how the adjustable parameters in the model are tuned against chamber observations, and as a result the enhancement in SOA production in the absence of vapor wall loss could vary with different model parameterizations. The authors mentioned that the OA concentrations from FLAME-III experiments are used to constrain the model performance, yet the comparison of simulations with experimental observations are not given in details throughout of the paper. Ideally, the organic aerosol temporal profile by AMS/SMPS during one representative experiment should be given together with corresponding simulations (e.g., Figure 3) to better visualize the model performance. Another question related, have the authors conducted optimal fitting of simulations to chamber measured quantities such as organic aerosol mass, O:C and H:C ratios? Is there more than one set of parameters that could well represent the observations? What is the physical meaning of each best-fit parameter that is chosen to describe the BBOA evolution?

For the ambient plume simulations, the authors are suggested to add discussions on how the values of key parameters, such as fire sizes and atmospheric stability classes, are assigned. Are they representative of the fire plume transportation in the air? A thorough search on the ambient fire plume properties in literatures might be useful to rationalize the sensitivity tests conducted in this study.

Recent two-dimensional VBS frameworks have incorporated gas-phase fragmentation processes as a function of the O:C ratio of individual volatility bins (e.g., Jimenez et al. 2009). The original distribution of volatility bins upon one generation of oxidation (drops in volatility per reactions) would correspondingly change by adding this branch of mechanism into the model framework. Upon OH-exposure in the order of ~ 10^{10} molecules cm^{-3} s (typically several hours of reactions in the atmosphere), fragmentation should have occurred to some extent, depending on the OH reactivity of the parent precursors. The authors are suggested to discuss uncertainties caused by the assumption of zero fragmentation in the reaction mechanisms.

Minor:

Page 5, Line 168: How are the black carbon and organic contents treated in each particle size bin in the model? Are they well mixed?

Page 9, Line 328: Add ‘are’ before ‘shown’.

Page 29, Figure 1: How are the vapor concentrations calculated, based on equilibrium partitioning?