

***Interactive comment on* “Laboratory investigation of photochemical oxidation of organic aerosol from wood fires – Part 1: Measurement and simulation of organic aerosol evolution” by A. P. Grieshop et al.**

**Anonymous Referee #1**

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This paper summarizes the results of well-designed and comprehensive set of experiments which challenge the traditional classification of organic aerosols into primary biomass burning OA and SOA. The results are very important in both apportioning of organic aerosol sources and improving the approach taken in atmospheric chemistry transport models. The experimental conditions are set to mimic conditions prevalent in biomass smoke plumes, including the presence of primarily emitted organic aerosol particles, with the exception of plume dispersion (dilution) in a real atmosphere (page 15710 line 5). It is beyond any doubt that the results seriously undermine the con-

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servative view in atmospheric modeling that combustion derived POA are non-volatile whereas SOA are semi-volatile. However, the lack of dilution in the experiments raises some doubts to what extent the results can be adapted in chemical transport models in spite of the fact that their currently applied simplified approach is also not valid. The authors found a significant increase (by a factor of 1.8 to 2.7) in OA mass after 4 to 6 hours of photochemical aging. Atmospheric dilution would be expected to reduce this effect significantly due to a combination of several factors: 1) depletion of the pool of semi-volatile organic compounds by volatilization, as observed in other source dilution experiments (page 15715 line 1); 2) reduction of the reaction rates due to reduced concentrations of precursors; 3) depletion of the pool of freshly formed SOA by volatilization (although the authors found that SOA is substantially less volatile than POA, it still had a significant volatile fraction of 20-40 % by mass, page 15713 line 12). Taken together, it might well be that the atmospheric dilution effects would override the experimentally observed increase in OA in smoke plumes, leading to a net decrease in OA as the plume ages in the atmosphere. It may also mean that the oversimplified approach taken by chemical transport models (POA non-volatile, SOA semi-volatile) may not be as bad as indicated: the loss of POA is compensated by non-volatile SOA to an unknown extent. What may happen there is a chemical transformation of POA into a combination of POA+SOA, with a highly uncertain net effect on the mass concentration of OA. Therefore without considering the effects of dilution experimentally the direct comparison of experimental results with field observations (such as on page 15719-15720) may not be justified. Clearly, the paper represents a self-consistent and very important contribution to our understanding of basic processes undergoing during plume aging, but clearly further studies are need before this knowledge can be adapted in chemical transport models.

Minor comments: BC is considered as a conservative tracer in the experiments and used to compensate for wall losses in the experimental chamber. However, given that it was measured with an aethalometer and the optical properties of soot may change upon acquiring an organic coating or simply by physical aging, this may introduce some

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uncertainty into the correction approach which needs to be discussed.

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