Comment on acp-2021-380
Anonymous Referee #1

Referee comment on "Technical Note: Pyrolysis principles explain time-resolved organic aerosol release from biomass burning" by Mariam Fawaz et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-380-RC1, 2021

The paper addresses an important issue in understanding the reported variability of emission of organic aerosol (OA) from wood combustion which is not well constrained. Accurate measurement of emission factors (EF’s) is important since EF’s have long been the fundamental tool in developing national, regional, state, and local emissions inventories for air quality management decisions and in developing emissions control strategies. This work contributes to an effort to standardize the process in obtaining EF’s and a great first step in developing a predictive understanding of release of particles and gases from pyrolysis. The paper is well written and brief and concise and worthy of publications with some explanations.

Biomass combustion is a complex process that consists of consecutive heterogeneous and homogeneous reactions, and a single particle of the biomass combustion process; consisting of: – heating and drying, – pyrolysis (devolatilization), – flame combustion, and – char combustion. Each process is generally believed to depend on the biomass fuel type, geometry, temperature and the combustion environment.

General comments:

The factors controlling pyrolytic properties of biomass need to be discussed in greater detail: For example, the pyrolytic properties of biomass are controlled by the chemical composition of its major components, namely cellulose, hemicelluloses, and lignin and their minor components including extractives and inorganic materials. The fuel properties and process conditions affect the pyrolysis and combustion characteristics, altering the heat generation, heat transfer and reaction rates in a complicated manner See for example [Ryu, C., et al., Effect of Fuel Properties on Biomass Combustion: Part I, Experiments â Fuel Type, Equivalence Ratio and Particle Size, Fuel, 85 (2006), 7-8, pp. 1039-1046] For a biomass with higher cellulose content, the pyrolysis rate became faster, while, the biomass with higher lignin content gave a slower pyrolysis rate, which means that pyrolysis of cellulose and hemicellulose will proceed to completion before the pyrolysis of lignin reaches a very advanced stage [see: Gani, A., Naruse, I., Effect of Cellulose and Lignin Content on Pyrolysis and Combustion Characteristics for Several Types of Biomass, Renewable Energy, 32 (2007), 4, pp. 649-661, and Roberts, A. F., A Review of Kinetics Data for the Pyrolysis of Wood and Related Substances, Combustion and Flame, 14 (1970), 2, pp. 261-272]. The cellulose and lignin content in the biomasses
were one of the important parameters required to evaluate pyrolysis characteristics which suggests fuel type dependence contrary to the conclusions by the authors. 

Line 139: “constant temperature experiments at 400, 500, and 600 °C”. The distinction between slow and fast pyrolysis has been described for example 400 °C are considered as slow pyrolysis which is an irreversible process in which thermal decomposition of the biomass material and residence time varies from 5 to 30 min in this case biomass is slowly devolatilized, and volatiles do not escape as rapidly as they do in fast pyrolysis. On the other hand, 600 °C is considered fast pyrolysis and characterized with a short residence time, [ see: Vamvuka, D., Bio-oil, Solid and Gaseous Biofuels from Biomass Pyrolysis Processes â– An Overview, International Journal of Energy Research, 35 (2011), 10, pp. 835-862, Mohan, D., et al., Pyrolysis of Wood/Biomass for Bio-oil: A Critical Review, Energy & Fuels, 20 (2006), 3, pp. 848-889]. The authors need to clarify if their experiments correspond to fast or slow pyrolysis.

Line 297-299: “We have shown that differences in yields of particles and gases could be explained by reactor temperature, moisture content, and wood size, and that time-dependent emission of products can be explained by differing thermal diffusivities rather than wood species.”

It is certainly true that that wood particle pyrolysis process is dependent on its size, geometry and specific, anisotropic structure. It was shown that the chemical composition of biomass and its main constituents (lignin, cellulose, and hemicelluloses) plays an important role in the pyrolysis process. Thermal degradation of lignin begins at 200 °C and between 225 °C and 450 °C the reaction process becomes exothermic. Unless we assume that all wood samples have similar chemical composition the dependence on the wood species cannot be ignored. There are a lot of differences in the structures of hardwood and softwood, which has a large influence on the heat and mass transfer processes occurring during pyrolysis. Softwood (such as spruce) has long fibers (1.9-5.6 cm) and a more homogeneous structure, while hard wood (such as beech) has a heterogeneous structure and small fibers (0.4-1.9 cm) [Sokele, B., Wood Chemistry (in Bosnian), University of Sarajevo, Sarajevo, 197; Westerhof, R. J. M., et al., Effect of Particle Geometry and Microstructure on Fast Pyrolysis of Beech Wood, Energy Fuels, 26 (2012), 4, pp. 2274-2280]. These specifics have a strong influence on the diffusion mechanisms related to heat and species transfer trough biomass particle.

In fact, Petar M. GVERO et al Pyrolysis as a Key Process in Biomass Combustion … THERMAL SCIENCE: Year 2016, Vol. 20, No. 4, pp. 1209-1222, showed the importance of fuel and environment characteristics on the pyrolysis and combustion process of biomass. Regarding to mass transfer in the biomass particle during pyrolysis, two types of diffusion processes occur: diffusion of the bonded moisture through cell walls, and diffusion of the gas’s mixture. Diffusion is the dominant transport mechanism during pyrolysis that occurs on the lower heat fluxes and temperatures, while at higher temperatures, convection is dominant.

Additional Comments:

- Even though citation is provided for the experimental details a little more detail would be helpful.
- The range of temperatures selected for this experiment may not be representative
temperatures in wildfires or even cookstoves. What happens at much higher temperatures corresponding to flaming fires?

- Is there a way to relate and quantify the pyrolysis process to the commonly used MCE commonly used to determine combustion conditions?

Please also note the supplement to this comment: https://acp.copernicus.org/preprints/acp-2021-380/acp-2021-380-RC1-supplement.pdf