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Comment on acp-2021-930

Anonymous Referee #2

Referee comment on "Cellulose in atmospheric particulate matter at rural and urban sites across France and Switzerland" by Adam Brighty et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-930-RC2, 2021

The manuscript presents a comprehensive and valuable study on the atmospheric presence of cellulose based on an impressively large dataset. Cellulose is not the most critical species in atmospheric aerosol, yet a better understanding of its sources and atmospheric distribution is important in global aerosol and climate modelling. The work relies on the methodology of previous studies and in some respect contradicts to some of their findings. The sampling and analytical part of the study is scientifically sound, and the resulting dataset is reliable.

However, I have major reservations concerning emission source apportionment of cellulose based on correlation studies (sub-chapter 3.5). One of the findings of the authors is that biomass burning is not a source of cellulose for lack of correlation with the concentrations of the biomass burning tracer levoglucosan. I never understand if there is a robust methodology for determining cellulose from particulates, and a hypothesis that biomass burning might release plant debris, why not test it by carrying out combustion experiments and determine cellulose from sampled smoke particulates? Why do the authors believe that simple correlation can prove or disprove such a hypothesis? Let us imagine a plausible scenario that is tested with correlation studies. Assume that cellulose are released from biomass burning, but not from wood burning where cellulose is strongly bound into the wood matrix, but from the burning of garden waste, of partially decomposed leaves and twigs, from which the escape of unburned plant debris may be possible. Beyond any doubts, atmospheric concentrations of levoglucosan will be governed by wood burning, since in any environment the mass of firewood dominates all the biomass that is burned. In this case, though cellulose IS released by a process of biomass burning, there will be no correlation between the measured concentrations of cellulose and levoglucosan simply because the dominant process (i.e. wood burning) overrides the signal. In addition, cellulose and levoglucosan should not come from the very same combustion process since levoglucosan is the pyrolysis product of cellulose itself. Thus, the plain statement that “the sources of atmospheric plant debris do not include any significant input from biomass burning” simply does not follow from the lack of correlation or negative correlation whatsoever.

The same applies for EC correlated for proving resuspension sources of cellulose. Let resuspension be a source of atmospheric cellulose. Imagine that in a town it is raining for a week. Since there is traffic all week, EC concentrations will be measured but the concentration of cellulose will be zero simply because there is no resuspension under such circumstances. Can we conclude from the lack of correlation that resuspension is not a source of cellulose? Of course not. In addition, EC and cellulose are in different size ranges (fine vs coarse), are clearly from different sources (tailpipe vs resuspension), and are bound to different conditions (all weather vs. dry conditions). Why would anybody expect similar behaviour that is manifested in perfect correlation? Simple analysis of resuspendable urban PM10 for cellulose would decide. Thus, the part of the manuscript on source apportionment by simple correlation is totally unfounded.