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Interactive comment on "Evaluation of the impact of wood combustion on benzo(a)pyrene concentrations, using ambient air measurements and dispersion modelling in Helsinki, Finland" *by* Heidi Hellén et al.

Anonymous Referee #1

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Review of the manuscript entitled: "Evaluation of impact of wood combustion on benzo(a)pyrene concentrations, using ambient air measurements and dispersion modelling in Helsinki, Finland" by Hellén et al. report medium yearly and monthly concentrations of benzo(a)pyrene and uses these concentrations and inventory data from a questionnaire and emission inventory in a simple dispersion model in order to evaluate the influence of wood burning in sub-urban areas in the Helsinki Metropolitan Area (HMA).

General Comments. In a pre-review of the manuscript it was already mentioned that

many variables, such as emission strength of sources, meteorology and chemical properties of analyzed compound are not well addressed and could cause uncertainties and discrepancies among predicted and measured data. However, the strength of the study is situated in the relationship between the "wood-burning" marker; levoglucosan, and benzo(a)pyrene, and the selection of urban sites and sub-urban sites. Although the authors show the strong correlation between the compounds, it can not appropriately quantify the apportion to BaP form other sources.

An important part of the manuscript and results is based on a questionnaire and data from studies that have been published by the "Helsinki Region Environmental Services Authority" in Finnish and can not be consulted without knowledge of this language. It is not clear what the uncertainties are of the data, and this is also not included in the presented manuscript. Especially, when comparing the results of measured versus calculated values, it is important to mention these uncertainties in order to get an insight on the quality of the outcome. Nevertheless, the around 800 household that were asked on their use of wood combustion gave a clear idea about the importance of this activity. Moreover, there seems to be no doubt that wood burning is an important source for benzo(a)pyrene in the HMA, which on its term could be useful in the discussion of the influence of semi-urban / semi-rural areas on regional air quality, since biomass burning is promoted as an energy source in the European Union.

The applied model is suitable for the studied region, but could be given more discrepancies in areas which are exposed to multiple sources and where wood burning will not be so dominant.

It would have been interesting to show more monthly data of all D-sites as well as U-sites to get a clearer overview on the results and the relationship between benzo(a)pyrene and levoglucosan, especially in relationship with co-variables, such as meteorological data.

Besides these general comments, the following doubts should be clarified:

Specific comments. Introduction: Pg2.ln8. A reference is missing on the trends in PAH in others parts in EU. In fact Southern Sweden and Northern Finland are not the only regions/sites were PAH (or specific BaP) is not decreasing, and many of these areas face similar situations as in the present study; i.e. combustion of wood or / and coal. Comment this here.

Ln.28. The outcome of the predicted BaP concentrations for the studied area should be mentioned and discussed here. What did the model predict for the studied areas, and was this related to wood combustion?

Pg3. ln2. The studies mentioned here are based on BaP and levoglucosan (and other compounds) measurements, and indicate that there are areas in EU which have high apportion of wood combustion for PAH.

Ln.8 to 11. This part could be left out from the manuscript, since it deals with PM2.5 and not BaP.

Ln.11 to Ln15 "In the Helsinki. . .sauna stoves". This sentence should be removed since it deals with PM2.5.

Ln23. "The very few studies" dealing with the "quantitative effect of residential wood combustion on the ambient air concentrations of PAHs" should be mentioned here.

Ln24. What do the authors consider "reliable estimations of the spatial distribution and temporal variations"? How are these items addressed in the present study? In my view, the authors present many sampling sites, and many sampling years, but few sites are sampled every year. This result is a mix of data that may not improve the reliability of the outcome. Discuss this in the manuscript.

Ln27. The authors could rewrite the sentence to: "Wood combustion is a major source of PAH (Shen et al. 2013), although the emission rates depend heavily on a large variety of factors, such as . . ."

Pg4.Ln2. The "new inventory" should be discussed and compared to "old" ones.

Ln3. There is a reference missing for the levoglucosan analysis in ambient air PM. Moreover it is not clear to me why the data of black carbon was not used in the present study, since this measured in the emissions (Gröndahl et al. 2011) in considerable amount (Savolhathi, et al. 2016). The used model should be introduced here.

Methods: Ln17. There should be a reference or measurements that demonstrate the "minor impact on air quality".

Measurements methods: Pg5.ln14. What is the uncertainty of the analysis at 0.200 ng/m3, and what do the authors mean with "estimated" measurement uncertainty? Please, clarify.

Ln10. Why do you want to pool samples? This will reduce the information of the sample day. Why was this done?

Ln20. It is not clear from the text if the samples of PAH and levoglucosan were the same filter, or different filter samples. It is also not clear if the samples for levoglucosan and PAH were collected on the same day and site. This should be clarified here and the sampling strategy should be discussed.

Ln25. It is not clear to me why the authors do not want to evaluate the emission from traffic. They use arguments, but here they have the tools to quantify the contribution. Please, clarify, why you do not want to quantify this contribution.

Ln31 (but also other issues mentioned from on pg.6 to pg7 referring to Kaski 2016). The report should be explained here in more detail, since this inventory is the fundament of your results. The mentioned report for more details is in Finnish, and many people do not control this language. It is also mentioned here that the black carbon emissions were estimated. It would have of major interest to show the results of BC measurements in relation to BaP and other tracers for the apportion of wood combustion (and other sources) on the ambient air.

Pg6. Ln1. What is the influence of these factors on the results in this study? Explain in more detail.

Ln31. What are the uncertainties of these factors in the present study? Is it possible to introduce them in the final result, so the reader understands the error of the calculations and will be able to validate the model better?

Pg7.ln12. What do you mean you did not get enough information form the questionnaire to estimate the influences of meteorological variables on the emissions? These variables have influence on emissions (also see pag.10.ln30 and pg11.ln27). Clarify the reliability the questionnaire.

Atmospheric dispersion modelling: Pg7.ln21. It is not clear why the model was run on an hourly base while BaP levels are monthly concentrations. Please, clarify.

Pg8.ln6. Why was particle bounded BaP treated as an inert gas?...why not as an inert particle, or a reactive particle? Discuss the differences between these possibilities and the influence on the outcome of the model.

Correlations of the concentrations of BaP and levoglucosan Pg9.ln23. Here it is mentioned that levoglucosan may not be a quantitative tracer due to its reactivity and dependence of combustion conditions, but this could be compared to BaP, which exhibit similar properties. Are they comparable?

Pg.10.ln1. The observed ratio in the present study should be compared to more than one study (Belis et al, 2011). In fact, the Belis study is also based on measurements, like the present study. The observed difference of a factor 10 should therefore be discussed in other terms. It is important here. For your interest; Fine et al. 2004. ENVIRONMENTAL ENGINEERING SCIENCE 21. observed BaP to levoglucosan ratios closer to 0.001 then 0.01. Why was the relationship between BaP and levoglucosan not used to "estimate" the contribution of wood combustion throughout the year and daily, as was done elsewhere (see Belis et al, 2011, or van Drooge & Perez Ballesta. 2009. ENVIRONMENTAL SCIENCE AND TECHNOLOGY.43.7310)?

Predicted spatial concentration distributions: Pg10.ln24 (and second paragraph). Is not clear how the "differences in meteorological conditions" influence the high and low BaP levels. What are these conditions and how are they different. Please, clarify.

Pg11.ln6 to 12. The comparison to the Czech study is almost irrelevant, since this is another situation, other sampling method and traffic included-model. The comparison could be removed from the manuscript. Are there no other studied to compare, and what would happen with the model outcome if traffic emissions were included?

Comparison of the observed and predicted average annual concentrations. It is not clear why only annual results are compared? Why not monthly results, or at higher temporal resolution. It is interesting to see how the BaP concentrations fluctuate along the year in the different months (or weeks, or days, such as weekend versus weekdays, in the HMA) It is unclear why 0.135 ng/m3 was add to the "computed concentration". If this is background, where does it come from? It is a considerable level. Why the regional background from Hyytiaäla was used and not a regional urban background from a urban background station, or urban site with low levels (as observed in this study)?

Ln.27 and whole paragraph. It is not clear why the temporal variation of the emissions were not addressed better. If this emissions are based on daily to monthly variations (not really clear how), it is not clear why this was not possible to investigate the influence of the meteorological conditions on the emissions.

The authors declare that many factors, such as meteorological influences on emissions, reactivity of BaP, particle-bounded properties of BaP, and the use of a regional background in the vicinity of the studied area were not taken in to consideration when they started the modelling, but these factors are well known beforehand. Can the authors improve their model? Really, mentioning these limitations in the last part of the discussion is not appropriate. These questions are clear from the beginning and should have been introduced right from the start, or the fact that they were not introduced (as was the case here) they should have been mentioned and discussed beforehand. The model is probably not that bad, but misses a clear uncertainty calculation which makes the discussion too speculative.

I recommend to authors to focus on the real results, the chemical analysis that show that the sub-urban (detached house) zone is an area of seriously high BaP concentrations which is link directly to wood burning based on levoglucosan levels. This wood burning is confirmed by the habitants of the area which declare in the questionnaire that they use wood for (secondary heating) and saunas. These saunas are probably typical in Nordic countries, while in other areas domestic heating is more important. It does not matter. The results from the questionnaire are used in a simple model to see if this wood burning coincides with the measured BaP concentrations. They do, but it is not clear to what extend..since there is an error calculation missing.

The authors could mention that the applications of questionnaires in the case of wood burning are very powerful, since a national/regional inventory on wood burning is not existing or underestimating the real wood consumption, since the wood is often non-invoiced and self-supplying wood. In the context of the present study a comparison with other questionnaires could be made, such as the one by Pastorello et al. 2011. ATMOSPHERIC ENVIRONMENT. 45.2869–2876.

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